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THE AMERICAN MINERALOGIST

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MINERAL FORMATION IN NATURAL CHEMICAL SYSTEMS UNDER EQUILIBRIUM AND NON-EQUILIBRIUM CONDITIONS*

GEORGE TUNELL, *University of California, Los Angeles, California.*

The processes of formation of minerals can be observed while they are going on in some cases; in most cases, however, the processes ceased before the mineral assemblages were exposed to the view of the mineralogist and geologist, and the reactions that took place can only be inferred from the nature of the mineral assemblages that they produced.

Minerals are formed in nature in many ways. They crystallize slowly from magmas deep in the earth's crust. They precipitate from hot ascending solutions. They result from the interaction of hot ascending fluids with the rocks penetrated by the fluids. The congelation of lavas poured out on the earth's surface produces mineral aggregates in some cases and volcanic glass in others. Air and rain water act upon the minerals at the earth's surface and penetrate some distance below it decomposing many minerals and carrying away part in solution. These solutions react with some of the minerals in the rocks through which they pass giving rise to additional minerals. Some of the solutions again emerge as springs and join the waters flowing on the surface in streams and rivers in which dissolved materials are carried to lakes or to the sea. From lakes and seas still more minerals are precipitated.

The crystallization of magmas in intrusive bodies proceeds slowly because the latent heat of crystallization is conducted away slowly by the overlying rocks. Laboratory investigation of many silicate systems has shown that the rate of establishment of equilibrium at liquidus temperatures is of the right order of magnitude so that in all probability approximate equilibrium is maintained between the magma of intrusive bodies and the minerals crystallizing from it at the time they crystallize. Minerals that have crystallized out in some cases react with the magma later on when they are no longer stable in contact with it; in other cases they persist unchanged, although with further cooling of the magma they

* Address of the retiring President of the Mineralogical Society of America given at the thirty-first annual meeting of the Society in Washington, D. C., on November 16, 1950.

are no longer in equilibrium with it and should change in composition or be redissolved. Incompletely resorbed crystals of olivine and zoned crystals of feldspar and pyroxene in many rocks indicate that the minerals did not maintain perfect equilibrium with the magma after their formation.

Insofar as the crystallization of a particular portion of magma proceeds with approximate maintenance of equilibrium between the magma and the crystals forming from it, the process can be represented approximately by a line or curve in an equilibrium diagram. If some of the crystals are removed from the portion of the magma under consideration by sinking or floating, the further course of crystallization of the remaining portion can still be represented by a line or curve in an equilibrium diagram, but plotting the further course of crystallization is only possible if the mass of the crystals removed is known as well as their composition. The use of equilibrium diagrams to portray processes in which the mass of the system is changing is possible because the changes of state of a system of variable mass are represented by a definite path in the equilibrium diagram provided the mass subtracted or added is in equilibrium with the solution at the time it is subtracted or added. If crystals once formed become unstable in relation to the magma when the temperature falls, but fail to react with the magma because they are separated from it by a protective coating, it would be necessary to know the mass and composition of the crystals so separated in order to plot precisely the further course of crystallization of the remaining magma. Without knowledge of the mass of crystals removed from the magma or separated from it by protective coatings it would still be possible, however, to tell the general nature of the deviation of the further course of crystallization from the path that would have been taken if no crystals had been removed or separated by protective coatings, provided the composition of the crystals removed from the magma or separated from it is known.

As a result of investigations of equilibrium relations in silicate systems it has become generally accepted that the larger scale field and smaller scale microscopic relations of the igneous rocks are in many cases interpretable as the results of crystallization during which the reaction relations of many of the minerals and the differing extents to which equilibrium was maintained brought about a diversity of mineral assemblage in the intrusive igneous rocks even from initially similar magmas.

Liquid magma poured out on the earth's surface as lava from fissures or vents in some cases cooled slowly enough so that it became completely crystalline, but in other cases it congealed to a mixture of crystals and glass; in still other cases lava masses solidified as almost completely glassy rocks. The series of states through which the magma passed as it changed from a liquid to a glass were non-equilibrium states and the process cannot be represented by a curve in an equilibrium diagram.

During the crystallization of magmas in deep seated intrusions the residual fluid became enriched in water and other constituents not present or present to a very limited extent in the early formed crystalline phases. The resulting fluids in part crystallized within the intrusive bodies where they formed pegmatitic masses grading into the surrounding rock of the intrusive. In part the residual fluids were forced into the rocks surrounding the intrusive along fissures or planes of weakness and there formed pegmatite dikes. During the crystallization of the pegmatitic masses and dikes the minerals crystallizing out of the fluid may well have been in approximate equilibrium with the fluid at the time they crystallized; the large crystals found in pegmatites are regarded as indications that growth proceeded for a long time without large changes of temperature, pressure or concentration. Moreover, it has been found in laboratory experiments with related systems of silicates and water that equilibrium is rather rapidly established at temperatures and pressures near those that obtained during the crystallization of pegmatites. As the character of the solutions changed, however, some of the early formed minerals became unstable and were replaced by others, but the alterations in the crystalline phases probably did not always keep pace with the changes in the fluid phases. Some minerals although unstable have persisted in part until exposure of the pegmatitic masses and dikes by erosion.

Adams¹ has stated that for an understanding of pegmatite formation data on the equilibrium relations in the very complex system $\text{Na}_2\text{O}-\text{K}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$ at temperatures below about 600°C are needed. Although data for this quinary system are not yet available, investigations of the equilibrium relations in the systems $\text{Na}_2\text{O}-\text{SiO}_2-\text{H}_2\text{O}$ and $\text{K}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$ are being conducted at the Geophysical Laboratory and it is hoped will lead to a determination of the equilibrium relations in the part of the quinary system that is of interest for its application to pegmatite genesis. Data for the ternary systems $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$ and $\text{K}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$, which are parts of the quinary system, were published by Schairer and Bowen² several years ago, and very recently the results of the comprehensive study of the system $\text{NaAlSi}_3\text{O}_8-\text{KAlSi}_3\text{O}_8-\text{H}_2\text{O}$ by Bowen and Tuttle³ appeared. At high temperatures soda feldspar and potash feldspar form a continuous series of solid solutions of the type with a minimum melting temperature at an intermediate composition in the series.⁴ The diagram is complicated toward the potash-rich side by the incongruent relations resulting from the

¹ Adams, L. H., Carnegie Institution of Washington Yearbook, No. 38 (1939) page 38.

² Schairer, J. F., and Bowen, N. L., *Am. Jour. Sci.*, **245**, 193-204 (1947).

³ Bowen, N. L., and Tuttle, O. F., *Jour. Geol.*, **58**, 489-511 (1950).

⁴ Schairer, J. F., *Jour. Geol.*, **58**, 512-517 (1950).

separation of leucite. In the presence of water vapor under pressure Bowen and Tuttle found that the melting temperatures are lowered substantially and they determined the precise values of the lowering at several pressures. At temperatures below the melting curve at the pressures used the homogenous feldspar that crystallized from the melts unmixes, a relation that was deduced from study of natural perthites by previous investigators. The actual position of the unmixing curve or solvus was determined by Bowen and Tuttle. Unlike the liquidus and solidus curves, the solvus does not change its position measurably with change of pressure of water vapor because water does not participate in the equilibrium at the solvus.

Other residual magmatic fluids deposited metallic ores with accompanying gangue minerals in the adjacent and overlying rocks as well as in parts of the magma already congealed. The composition and state of these fluids in depth can only be inferred from the nature of the deposits that precipitated from them or resulted from their interaction with the rocks invaded by them. Careful field and microscopic study of deposits formed in depth has yielded some knowledge of the chemical elements carried by the ore-forming fluids. From observations of the structures and textures of the deposits and of the minerals formed, various suggestions have been made by ore geologists as to the physical character and chemical composition of the ore-forming fluids, but considerable uncertainty and difference of opinion remain on these points. That the attack on the difficult problems of ore genesis should begin with careful and accurate mapping and recording of the geologic features of ore deposits, including structures, textures, mineral associations and alteration of enclosing or adjacent rocks, has been emphasized by numerous investigators and seems beyond dispute. Mining geologists are concerned most of all with the structural features of ore deposits, since the location, size and shape of ore bodies depend so much on structural factors. However, such experienced and eminent ore geologists as Butler, Hewett, Sales, Locke, Ransome, Gratton and Blanchard have concluded that field studies need to be supplemented by laboratory investigations in order that a better understanding may be obtained of the chemical aspects of ore deposition.

A number of studies of solubilities of ore minerals have been carried out in recent years and have yielded some valuable information. The importance of determinations of sulfide solubilities in systems containing silica and various other components that are believed to have been present in the ore-forming solutions (since quartz and various other gangue materials were deposited along with the sulfides) is undoubtedly great. It appears that in order to obtain an understanding of the ore

mineral associations, systematic determinations of the phase equilibria in a number of systems made up of the constituents of ores plus water, H_2S , H_2SO_4 , CO_2 , Cl_2 , F_2 , etc., should be undertaken. It might then be possible to deduce from the equilibrium diagrams of such systems some of the series of minerals that would be precipitated by the cooling of solutions in such systems or by their reaction with wall rocks or by the lowering of external pressure. If a particular mineral association and sequence could be shown to result from one of these solutions by cooling, reaction or pressure release, we would then have stronger evidence than is now available that the hypothetical solution was approximately like the actual ore solution responsible for the natural ore deposit. The needed equilibrium investigations, although difficult and laborious, do not appear to be entirely beyond the range of present technical resources. The determination of homogeneous equilibria, that is, the molecular and ionic groupings in the solutions under equilibrium conditions, would not necessarily have to be carried out. The determination of the heterogeneous equilibria alone, that is, the compositions of the fluid and crystalline phases in equilibrium at the various temperatures and pressures in certain systems might be found sufficient to throw considerable light on the conditions of formation of the ore minerals and the nature of the ore fluids. The system hydrogen-oxygen-sulfur and the system hydrogen-oxygen-sulfur-iron are two that were recommended for investigation by the National Research Council Committee on Problems of Ore Deposits⁵ in 1940 as fundamental to the further understanding of the chemistry of ore deposition. The equilibrium of the solid phases and vapor phase in the system copper-iron-sulfur was the subject of a comprehensive study by Merwin and Lombard.⁶ Their work was carried out at a series of vapor pressures up to the melting curve. If the heterogeneous equilibria in the system hydrogen-oxygen-sulfur-iron were determined in the temperature-pressure-concentration range relevant to ore formation problems, the results of Merwin and Lombard would then provide a basis for extending the equilibrium data to the system hydrogen-oxygen-sulfur-iron-copper in the relevant temperature-pressure-concentration range. Knowledge of the equilibrium relations in this system would certainly be of great interest for its bearing on the hypogene mineralization of the porphyry copper deposits.

The quicksilver deposits are among the hydrothermal deposits believed to have formed at relatively low temperatures and pressures. They have been carefully studied in the field and their structural and mineralogical

⁵ Report of the Committee on Problems of Ore Deposits, National Research Council, Washington, D. C. (1940) (Mimeographed Draft).

⁶ Merwin, H. E., and Lombard, R. H., *Econ. Geol.*, **32**, 203-284 (1937).

features have been well described. The simplicity of the mineralogy and the probability that they were formed at shallow depths by solutions not very different from hot spring waters such as those now emerging at Steamboat Springs, Nevada, should make it possible to deduce the main features of the processes by which they originated, with less difficulty than would be the case with most metalliferous deposits. According to Brannock, Fix, Gianella and White,⁷ at Steamboat Springs the hot waters are now depositing fine grained dark-gray to black mud at the surface, consisting mainly of particles believed to be silica gel or opal and containing stibnite and pyrite and appreciable amounts of gold, silver, mercury and antimony in undetermined form. A large quantity of originally porous sinter at Steamboat Springs that is now buried under younger sinter has been reworked by the hot waters after burial and filled with opal, chalcedony, and some quartz. Radial clusters of stibnite occur in small cavities and embedded in opal in the reworked sinter. Fine-grained cinnabar also is found in many parts of the reworked sinter.

Analyses of the waters of the springs indicate a parent saline magmatic water diluted to varying degrees by meteoric water of low concentration. The water in the different springs at Steamboat Springs ranges from slightly acid to moderately alkaline and on the average is slightly alkaline. The acidity or alkalinity of the water appears to be related to the temperature of the spring, with the most alkaline waters tending to be those issuing at the highest temperatures.

In the Goldbanks District of Nevada a commercially productive cinnabar deposit was formed syngenetically with a silica apron deposited by thermal springs, according to Dreyer.⁸ Many of the opalitic cinnabar deposits are stated by C. P. Ross⁹ to have been formed within a few score or at most a few hundred feet of the surface, and in a few, such as those of the Coso District of California, the cinnabar crystallized in hot springs at the surface. Many other types of quicksilver deposit have been described in the United States, but all appear to have been formed close to the surface and at relatively low temperatures.

It has long been known that mercury sulfide is appreciably soluble in alkaline sulfide solutions. The solubilities of cinnabar were determined by Knox¹⁰ for a range of Na_2S concentrations at 25° C and 33° C. Knox's equilibrium investigations showed that an increase in the concentration of Na_2S in the solution causes an increase in solubility of cinnabar in the

⁷ Brannock, W. W., Fix, P. F., Gianella, V. P., and White, D. E., *Trans. Am. Geophys. Union*, **29**, 211-226 (1948).

⁸ Dreyer, R. M., *Econ. Geol.*, **35**, 141-145 (1940).

⁹ Ross, C. P., *Econ. Geol.*, **37**, 461 (1942).

¹⁰ Knox, J., *Zeit. f. Elektroch.*, **12**, 477-481 (1906).

system $\text{HgS-Na}_2\text{S-H}_2\text{O}$. That the mercury sulfide of the quicksilver deposits was transported in alkaline sulfide solution has been generally accepted by geologists who have studied these ores. There is not such complete agreement, however, as to the relative importance of several possible causes of the deposition of the cinnabar. The causes to which the precipitation of the cinnabar has been ascribed are chiefly evaporation of solution into open spaces in the wall rock, reaction of the solution with the wall rock, reduction of the alkalinity of the solutions by admixture of acid meteoric waters, dilution by neutral meteoric waters, cooling of the hypogene solutions, and decrease of pressure. From Knox's data it may be concluded that dilution and evaporation are probably among the less important of these factors. Some quicksilver deposits contain metacinnabar as well as cinnabar, but others are devoid of metacinnabar. Allen, Crenshaw and Merwin¹¹ showed that metacinnabar precipitates from acid solutions under certain conditions. Its presence in some of the quicksilver deposits can be ascribed to acidification resulting from admixture of acid meteoric waters and/or to the oxidation of H_2S of the hypogene solutions by air oxygen close to the surface. Allen, Crenshaw and Merwin proved that cinnabar is the stable modification of mercuric sulfide over the whole temperature range up to the volatilization point (at atmospheric pressure). The deposition of metacinnabar in some quicksilver ores is thus an example of metastable phase formation. At the time it precipitated from the ore solutions the metacinnabar was probably in metastable equilibrium with those solutions. In some of the ores in which it has been found the metacinnabar has partly changed into cinnabar according to Dreyer,¹² probably as the result of an influx of additional hypogene alkaline sulfide solution.

In numerous quicksilver deposits the cinnabar is associated with silicified gangue; the silicification is in part much older than the cinnabar; in part the silicification and cinnabar deposition went on together as indicated by the very fine dispersion of much of the cinnabar in the quartz and chalcedony. The association of cinnabar with quartz, chalcedony and opal can be understood in a general way, since silica is appreciably soluble in solutions containing alkaline sulfide of mercury. Investigation of the equilibrium relations in a certain temperature-pressure-composition range in the system $\text{HgS-Na}_2\text{S-Na}_2\text{O-SiO}_2\text{-H}_2\text{O}$ might provide the basis for a more detailed understanding of the paragenetic relations. Knox's¹³ determination of the saturation surface of cinnabar in the system $\text{HgS-Na}_2\text{S-Na}_2\text{O-H}_2\text{O}$ would provide a starting

¹¹ Allen, E. T., Crenshaw, J. L., and Merwin, H. E., *Am. Jour. Sci.*, **34**, 341-396 (1912).

¹² Dreyer, R. M., *Econ. Geol.*, **35**, 149 (1940).

¹³ *Op. cit.*

point for the investigation of the equilibrium relations in the relevant part of the system $\text{HgS-Na}_2\text{S-Na}_2\text{O-SiO}_2\text{-H}_2\text{O}$. The association of cinnabar with other minerals, calcite, dolomite, pyrite, marcasite, kaolin group minerals, etc., is probably not as close in the main as its association with silica minerals. Some chemical aspects of these other associations have been discussed by several investigators, but not from the point of view of heterogeneous equilibria, necessary experimental data for such discussion not having been determined.

When ore deposits become exposed by erosion they are attacked by meteoric waters and atmospheric oxygen and carbon dioxide. At the surface and downward for a distance varying in different deposits the sulfide minerals usually are oxidized. Observation of oxidation products in outcrops impels the mining geologist to ask from what minerals were they formed and how much of the unoxidized minerals exists below. The importance of oxidation products in outcrops as possible indicators of unoxidized ore beneath has led to field and laboratory investigations aimed at better understanding of the oxidation of the sulfide minerals.

The disseminated copper deposits of the southwestern United States are relatively simple in mineral make-up. They consist chiefly of finely distributed pyrite, chalcopyrite, chalcocite and covellite in altered monzonite porphyry or altered granite porphyry or altered schist. As a result of the alteration of the original igneous or metamorphic rocks associated with the introduction of the copper, the ore bodies are crackled masses of fine-grained quartz and sericite together with fine-grained biotite and alunite and kaolin group minerals, through which the sulfides are disseminated in veinlets and in individual grains replacing the rock between the finest veinlets. The oxidized cappings above the ore bodies contain hematite, goethite, jarosite and nontronite in the veinlets and spaces where the sulfide minerals were before oxidation; the hematite, goethite and jarosite also are scattered through the surrounding rock as replacements of the sericite, kaolin group minerals and quartz. In some places important quantities of azurite, malachite and chrysocolla remain in the cappings, but in most places the copper has been leached from the cappings and carried downward in solution.

The reasons why iron minerals and copper minerals are in some places deposited in the cappings during oxidation whereas in other cases practically all of the copper and some of the iron are carried downward in solution are now fairly well established as a result of field and microscopic study together with laboratory determinations of the equilibrium relations in the relevant part of the system $\text{CuO-Fe}_2\text{O}_3\text{-SO}_3\text{-H}_2\text{O}$ ¹⁴ and

¹⁴ Tunell, G., and Posnjak, E. W., *Jour. Phys. Chem.*, **35**, 929-946 (1931).

supplementary experiments on equilibria¹⁵ and reaction velocities.¹⁶ The products formed and the quantities of each when given quantities of iron and copper sulfides are oxidized in a given quantity of water can be calculated from the equilibrium diagram of the system $\text{CuO-Fe}_2\text{O}_3\text{-SO}_3\text{-H}_2\text{O}$ provided all the iron be oxidized to the ferric state. It is noteworthy that the deposition of tenorite or brochantite or antlerite will only take

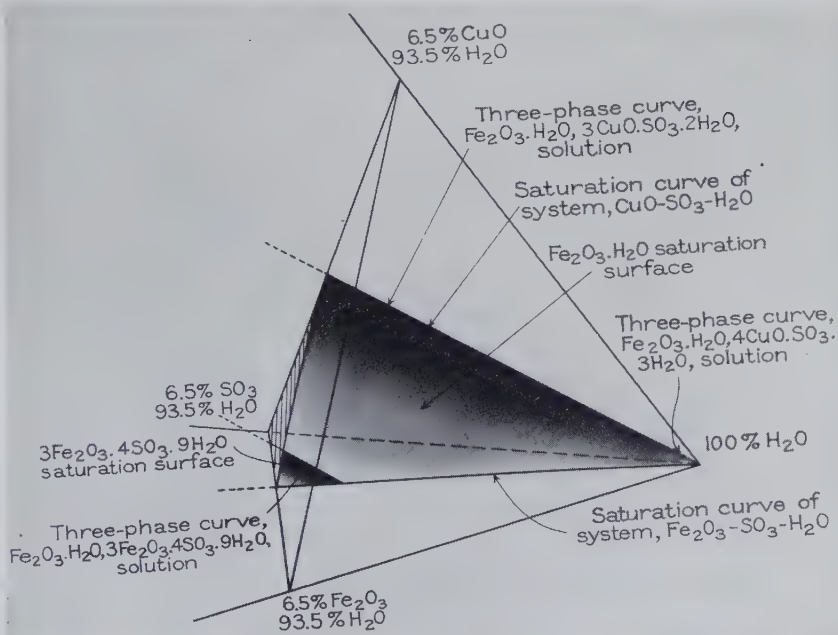


FIG. 1. Perspective view of a model of the isothermal-isobaric saturation surface in the portion of the system $\text{Fe}_2\text{O}_3\text{-CuO-SO}_3\text{-H}_2\text{O}$ in which the total composition is 93.5 per cent or more H_2O , 6.5 per cent or less Fe_2O_3 , 6.5 per cent or less CuO , and 6.5 per cent or less SO_3 .

place after practically all the iron (ferric) is precipitated as goethite or $3\text{Fe}_2\text{O}_3 \cdot 4\text{SO}_3 \cdot 9\text{H}_2\text{O}$. In order that all of the iron as well as all of the copper remain in solution a higher concentration of $\text{SO}_3(\text{H}_2\text{SO}_4)$ is required. In the oxidation of the disseminated copper ores the sericite is slightly attacked by the acid solutions. The small amount of potash taken into solution from the sericite plays an important role in the formation of hematite and jarosite as in its absence only goethite and

¹⁵ Posnjak, E., unpublished work.

¹⁶ Posnjak, E., *Amer. Inst. Min. and Met. Eng.*, Pamphlet No. 1615D (issued with *Min. and Met.*, December, 1926).

$3\text{Fe}_2\text{O}_3 \cdot 4\text{SO}_3 \cdot 9\text{H}_2\text{O}$ would be formed. That a very small concentration of potash causes the formation of jarosite in a solution that would otherwise precipitate $3\text{Fe}_2\text{O}_3 \cdot 4\text{SO}_3 \cdot 9\text{H}_2\text{O}$ was proved experimentally by Posnjak.^{17,18} The small amount of potash yielded by the decomposition of sericite is not sufficient, however, to cause the deposition of hematite where the total percentage of sulfides in the ore is moderate to high and where the ratio of pyrite to chalcocite and/or covellite is high. In the oxidation of such ore or protore goethite and jarosite precipitate. Oxidation of the disseminated copper ores appears to have been approximately thorough in some places, but in other places it was not thorough. The effect of non-thorough oxidation, in which some ferrous sulfate is present, is to diminish the quantities of goethite and/or jarosite precipitated, because there is less ferric sulfate and more sulfuric acid present. The knowledge gained of the chemical aspects of the oxidation process provides an explanation of the fact observed by Ransome¹⁹ that copper-stained rock traversed by many little veinlets of chrysocolla is abundant in the Miami District of Arizona, but as a rule the best ore bodies do not occur beneath rock of this character. Ransome inferred that the process of supergene enrichment is far more active when the material undergoing oxidation contains abundant pyrite than when it is chiefly chalcocite.²⁰ He observed that in the weathering of chalcocite, especially in feldspathic rocks, part of the copper is not carried downward as sulfate, but is converted into carbonate or silicate and remains near the surface. Ransome's observations of several features of oxidized cappings were very acute, and his conclusions concerning their significance in terms of underlying ore and waste were well founded and foreshadowed the results of later investigations.

I shall not proceed to discuss other types of mineral assemblage. I have attempted to indicate some of the controlling factors in the forma-

¹⁷ Unpublished work.

¹⁸ Jarosite and $3\text{Fe}_2\text{O}_3 \cdot 4\text{SO}_3 \cdot 9\text{H}_2\text{O}$ appear to have closely similar structures with one water molecule of $3\text{Fe}_2\text{O}_3 \cdot 4\text{SO}_3 \cdot 9\text{H}_2\text{O}$ taking the place of a potassium ion of jarosite and a second water molecule of $3\text{Fe}_2\text{O}_3 \cdot 4\text{SO}_3 \cdot 9\text{H}_2\text{O}$ taking the place of a hydroxyl ion of jarosite (S. B. Hendricks, *Am. Mineral.*, 22, 780 (1937)).

¹⁹ Ransome, F. L., *U. S. Geol. Surv., Prof. Paper No. 115*, p. 164 (1919).

²⁰ *Op. cit.* p. 171. (This conclusion of Ransome was an important advance in the understanding of leached outcrops. Ransome's comment on the same page regarding the statement of Zies, Allen and Merwin that sulfuric acid retards but does not change the nature of the reaction between pyrite and cupric sulfate resulted, however, from his failure to note that Zies, Allen and Merwin explicitly limited their discussion to the enrichment process alone as contrasted with the combined oxidation-solution-transportation-enrichment process to which Ransome applied their statement. The statement of Zies, Allen and Merwin is entirely correct in the case to which they clearly limited it.)

tion of just a few types. Field and microscopic investigations of the rock bodies and mineral deposits constituting the mineral assemblages discussed had inevitably raised the question: why are the particular minerals associated in the particular structures and textures? In some cases a partial answer to this question has been found through experimental determinations of heterogeneous equilibria and reaction velocities.

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HIGH TEMPERATURE THERMAL EFFECTS OF CLAY AND RELATED MATERIALS*

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ABSTRACT

A selected series of clay minerals and related silicates are examined by thermal, optical, and x -ray diffraction methods with a view toward establishing the significance of the various observed thermal effects. The heating range for the thermal curves is extended to 1300°C, and the study of specimens so fired is supplemented by the examination of the various materials after firing to several intermediate temperatures.

Detailed hypotheses are presented for the mechanisms of several of the structural changes observed as new phases are developed, and the influence of types of structures upon the nature and rapidity of such changes is discussed.

INTRODUCTION

The widespread interest in the use of thermal analyses for clays and related hydrous minerals has suggested the desirability of investigation of the nature of the various phase changes which cause the individual heat effects observed for the commoner types of materials to which thermal analyses are being applied.

Using the thermal curves as reference, the method of approach employed in this study has been simply to prepare portions of each of the specimens being investigated, fired to various intermediate temperatures below and above each prominent thermal effect, and to examine the resultant suites of specimens by x -ray diffraction and, where possible, by optical methods.

Initial developments of several of the high temperature phases are characterized by such small particle size that x -ray diffraction lines are noticeably broadened. In this condition particles are not resolvable microscopically, and one cannot even observe an average index for any single phase unmodified by refractivities of the matrix. In these circumstances, one is forced to rely entirely on the x -ray diagrams. It has further become apparent that not all of the materials encountered have been subjects of adequate prior description in the literature. To attempt to suitably characterize each of these somewhat variant phases is beyond the scope of this study. Instead, it seems desirable to interpose a short series of general remarks about each of the types which have not been found to be strictly correlative with standard literature descriptions—such as ASTM catalogues. Other phases, for which we are aware of no great latitude in diffraction characteristics, will simply be referred to without comment.

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THE PHASES

The three-layer anhydrides.

The altered diffraction effects observed from pyrophyllite after moderate ignition have been described by Thilo and Schünemann (1), and the observation of a similar phenomenon in the course of ignition of several montmorillonites and illites was previously remarked by the present authors (2). Consideration of the present and several other moderately fired montmorillonites suggests the following interpretation.

The removal of hydroxyl water, which is correlative with endothermal peaks around 500–700° and with increases of 0.1–0.3 Å in *c*-axis periodicities, involves the expulsion of about one-sixth of the oxygens of the octahedrally coordinated portion of the structure. This correlation is apparently confined to heptaphyllite type members.

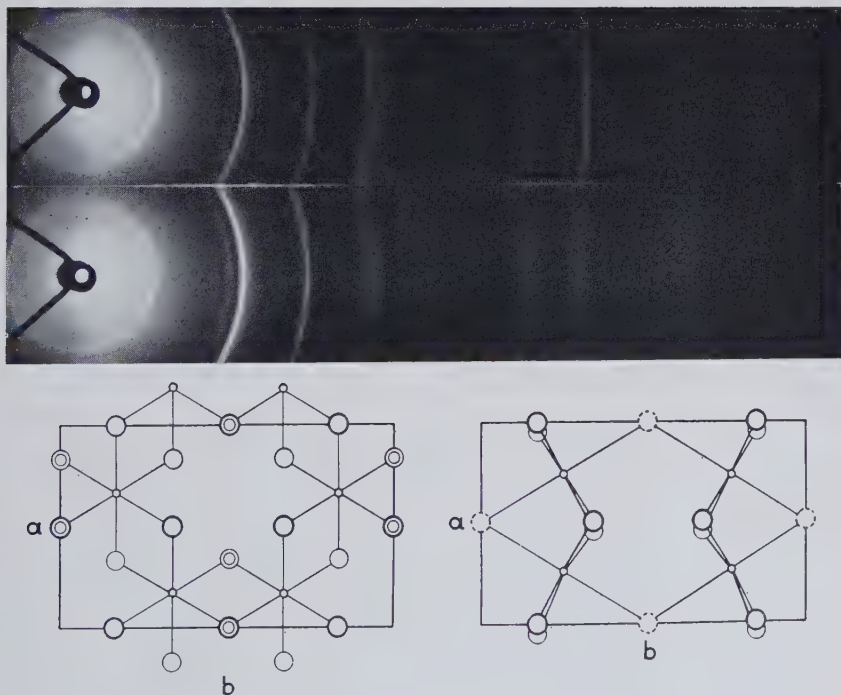


FIG. 1. The probable rearrangement in the heptaphyllite type clays upon loss of hydroxyl water. The diffraction diagrams illustrate the Tatatila montmorillonite after heating to 550° (upper) and 725° C. (lower). The schematic sketches represent the arrangement within an octahedral layer before and after dehydroxylation.

An idealized rearrangement of the octahedral layers which adequately accounts for the meager data observable is illustrated in Fig. 1. It

represents merely the lifting of the adjacent oxygen layers out of the packed position, with the immediate consequences that the vertical height is increased and the intensity of the 4.5 Å diffraction line is augmented. A diagram of a typical dehydroxylated specimen is compared with its normal dry state in Fig. 1.

TABLE 1. THE ANHYDRIDE OF THE TATATILA MONTMORILLONITE
IGNITED TO 725° C.

Indices as orthohexagonal	<i>d</i> in Å	Observed relative intensity	Calculated rela- tive intensity for idealized scheme of Fig. 1
001	9.7	m	2
002	4.85	m	2
110,020	4.48	ss	18
003	3.22	s	10
200,130	2.59	m	4
	2.5	diffuse m	
220,040	2.23	m	2
	2.1	diffuse w	
005	1.93	w	1
240,150	1.72	w	2
	1.67	diffuse m	
330,060	1.51	m	2
	1.49	diffuse w	
400,260	1.31	w	3
420,350	1.25	w	1

In Table 1 are summarized the diffraction data for the rearranged material. Prismatic reflections conform more nearly to hexagonal relations, their relative intensities are clearly altered, and in addition several diffuse scattering maxima are apparent at positions where better crystallized related minerals have strong pyramidal reflections.

The idealized scheme of Fig. 1 involves shared pairs of octahedral coordination faces for a heptaphyllite, and is probably unstable. Application of the same scheme to an octaphyllite type would demand sharing of three such pairs of faces, presumably impossible. For the magnesian clay from Hector, and for talc, the only two certain octaphyllites examined, no rearranged anhydride was observed.

THE SPINEL TYPE OXIDES

The spinel crystallization is well known, and it has even been possible to make useful correlations between composition and cell dimensions

within limited systems (3). Under the uncertain conditions of composition under which new phases develop in fired clays, however, the present authors have not attempted to search out any possible indication of composition. Indeed, as these spinel phases first appear, *x*-ray diffraction lines are notably diffuse and it is probably more true that rather than having any particular composition, the phase is merely an irregularly constituted assemblage of small cations, some octahedrally coordinated and others tetrahedrally coordinated, with only the oxygen packing actually approaching crystalline regularity. These spinels are not particularly refractory, however, and they grow rapidly to a clearly crystalline powder affording a clean diffraction diagram. No noticeable difference in relative intensities is observed between the diffuse and the sharp patterns, although cube edge parameters may change somewhat (with changing composition), and no erratic appearances of new lines have been remarked.

The character of the spinel type mixed oxides is thus in marked contrast to that of $\gamma\text{-Al}_2\text{O}_3$, a phase which has from time to time been described as a spinel-like structure. $\gamma\text{-Al}_2\text{O}_3$ also is probably some mixed arrangement of tetrahedrally and octahedrally coordinated aluminum ions, but the clean spinel type alumina material has been obtained only in the presence of a lithium mineralizer, and this clean material is actually a lithium aluminate.

The $\gamma\text{-Al}_2\text{O}_3$ normally observed upon ignition of gibbsite or boehmite varies markedly in character with the temperature of ignition at which it is formed (4), and the variations are much more profound than mere particle growth. Additional lines appear erratically, and probably of greater significance, the degree of diffuseness is unequal for different sets of lines, thus suggesting a distinct anisotropy, at least in the habit of the crystallites, and probably in the actual structure.

The apparent significance of these structural implications will be referred to again under discussion of the energy relationships inferred from the thermal curves for several individual specimens.

QUARTZ

Quartz has traditionally been looked upon as a phase of relatively fixed composition and invariant behavior. The cryptocrystalline synthetic products which have been found to result from firing of many montmorillonites at about 1000° C exhibit surprising vagaries with respect to their apparent relation to the high-low inversion. Synthesized quartz in the present study, when observed at room temperature, is found to exist in some cases as α -, in others as β -modification (in one case apparently intermediate).

In the absence of any criterion for the diffraction effects of the high form at room temperature, some attention must be given to the validity of its identification. Analyses of the material free from the matrix in which it is grown are not available. Either the α - or the β -quartz however makes the normal slow transition to cristobalite at more elevated temperatures.

Table 2 lists the observed reflections for the quartz which is developed in the montmorillonite from Otay, California, upon ignition to 1000° C.

TABLE 2. SYNTHETIC β -QUARTZ AT ROOM TEMPERATURE $a_0 = 5.11$ $c_0 = 5.37$ $u = .21$

Indices	d in Å	Observed relative intensity	Calculated relative intensity for powder diagram
100	4.43	m	20
101	3.42	sss	140
110	2.55	m	9
111		—	.2
102	2.30	w	2
200	2.22	m	4
201	2.045	m	4
112	1.85	ss	30
202	1.71	w	2
121	1.57	s	10
122 } 301 }	1.421	s	8 1.5
203	1.393	s	11
302	1.292	m	5
104		—	.5
220	1.277	m	5
123	1.225	w	4
131	1.196	m	5
114	1.190	w	3
132	1.113	ww	2
400	1.105	ww	1
105 } 223 }	1.044	ww	2 1

The observed a_0 and c_0 values represent reasonable extrapolations of the thermal expansion data of Jay back to room temperature (5). The apparent best estimate of the oxygen parameter is based mainly on the near equality of 121 and 122. It is probably sufficiently reliable to interpret in terms of dimensions within the structure. It is thus shown that

the O-O approach of the unbonded oxygen atoms across the 00 l open channels increases from 3.46 to 3.73 Å with falling temperature. An obvious explanation of the failure to invert would be the trapping of occasional extraneous ions in these channels.

Recent analyses of lithium aluminum silicates would suggest that lithium could be such an ion. Magnesium also is of suitable size. Neutrality would have to be maintained by inclusion of some aluminum.

CRISTOBALITE

Carefully prepared low-cristobalite is apparently a normal, well-organized crystal. The fine-grained manifestation resulting from syntheses in heterogeneous matrices are however noticeably variable (possibly even in composition (6)) and of uncertain inversion character. In addition there exist the opaline silica materials which are clearly sub-crystalline, but seem to be based upon the cristobalite crystallization. Such materials gradually become typical cristobalite without any important sudden energy effects upon ignition to sufficiently high temperatures. In the discussion to follow, no attempt is made to evaluate the quality of a cristobalite. The point is merely made that it is unlikely that poorly organized manifestations of such a crystal would be involved in any of the striking energy effects which provide the major features in a thermal analysis curve. The apparently abnormal instances of rapid evolution of cristobalite will be referred to later.

MULLITE

Mullite powder diffraction diagrams have been studied in meticulous detail by Rooksby (7), but the data are not correlated with the actual crystallization. It is probable that the sillimanite structure should be regarded only as a schematic representation, from which the pertinent observation to be made is that mullites are based upon centered chains of alumina coordination octahedra extending parallel to c and sharing two opposed edges. Details, such as the orientation of chains and the population of interchain cation positions are probably variable in moderate degree. No significance is attached to variations in crystals from separate sources in this study.

THE MAGNESIUM META-SILICATES

Studies of the preparation and polymorphism of magnesium meta-silicates (8) have indicated that the crystallization processes are probably too slow to provide energy effects identifiable in thermal analysis curves of already crystallized phases. It does appear to be true that the

crystallization of enstatite from amorphous hydrous magnesium silicates involves exothermal effects.

Enstatite and clinoenstatite are normal, well-crystallized phases and are observed in the firing of several high magnesian minerals. The MgSiO_3 which is formed in the firing of several typical montmorillonites is comparable with the "mesoenstatite" of Thilo and Rogge. This material is probably to be looked upon as a rudimentary nucleation rather than as a normal crystallization.

The structural relations of the layer silicates and the pyroxenes are such that projections onto the respective orthohexagonal axes are approximate equivalents. In the imperfectly developed diffraction diagrams of the fired montmorillonites the lines which appear are only those which relate to the pseudohexagonal nature. In fact, they constitute an identification of MgSiO_3 only in the sense that they correspond with equivalent lines which appear in better and better crystallized materials of higher magnesian content for gradations up to and including the typical enstatite crystallization obtainable from tremolite asbestos.

THERMO-CHEMICAL GENERALITIES

The method of thermal analysis is inherently an expression of the response of systems to nonequilibrium conditions. If thermal effects are to be apparent in the difference couple traces, it is required that reactions involving appreciable energy transfer be accomplished in a limited time. Endothermal reactions involving the crystallization of a new phase are of quite variable nature. Without going into quantitative aspects, which in most cases would be impossible for want of data, the following statements can be accepted. The rapid evolution of a hard, dense phase will afford a sharp exothermal effect. Sufficiently hard and dense crystals may, but need not, show broad exothermal effects in development over ranges of 50 to 100° C. Soft, irregularly constituted phases certainly cannot evolve detectable energy in slow development, and probably do not in moderately fast precipitation.

The simplest types of reactions, to which one would naturally like to look for standards by which to judge crystallization energy effects, do not present the features as resolved peaks. For example, for ignition of pairs like calcite and magnesite, where lime is softer than periclase, it is only apparent that following the decarbonation effect the return of the MgO temperature to the furnace temperature is more abrupt than that of the CaO . Similarly, for a set like brucite, hydrated lime and gibbsite, after dehydration the curves are respectively, abrupt, intermediate and gradual. For diasporite, which goes to the α - rather than the γ -oxide, the return is again abrupt.

The kind of reaction that does appear to produce a single sharp

exothermal effect identifiable separately on a curve is the kind for which substantial articulated units present in the structure of the starting material condense without significant rearrangement into the crystals of the new phase. Probably the clearest instance of this sort is the crystallization of forsterite ($H=6-7$, $G=3.21-3.33$) from clinocllore or from chrysotile. The latter case especially has attracted attention before, and can be discussed in some detail.

THE THERMAL SYNTHESIS OF FORSTERITE

The justification for projecting forsterite into a clay mineral study is that the syntheses may be accomplished from mineral specimens of far more perfect crystal development than that of the respective clay mineral analogues. Thus, there are available for study both chrysotile, which can be fired as a fibre, and high magnesium chlorites (clinocllore), which can be fired as cleavage lamellae. For each of these the crystal structures are known in sufficient detail to afford direct correlation between the natural phase and the synthetic forsterite. Each also exhibits an exothermal feature correlated with the development of the new phase.

Monochromatic pinhole diffraction diagrams taken through a fired flake of clinocllore and through a fired chrysotile fibre are reproduced in Fig. 2. Specialized orientation conditions for the new phase are readily apparent in each case. The chlorite flake affords three sets of strictly parallel grown forsterite aggregates mutually related to each other by rotations of 120° about a . The diagram amounts to three superposed preferred orientation diagrams each approximately along a for each of the three parallel grown sets. The latitude of deviation of a from the pole of the flake appears to be no greater than that normally introduced into flakes like chlorite or vermiculite by roasting them at lower temperatures where a layer silicate structure is preserved.

The chrysotile diagram, originally fibrous about c , now affords superposed forsterite rotation diagrams about the poles $[011]$, $[020]$, $[311]$, and $[320]$. The spots belonging to the $[011]$ and $[020]$ rotations are the clear, symmetrically shaped ones in layer lines of form, respectively $k+3l=0$, 1, 2, etc., and $k=0$, 1, 2, etc. Spots for the other rotations are weaker and are elongated parallel with the rotation axis. In Fig. 2 are illustrated the respective structural relations between the natural crystals and the fired product. It is seen that at the level of octahedrally coordinated Mg ions the transition from the old to the new phase requires no chaotic disintegration followed by reconstitution of a new assemblage, but simply the freedom of a few Mg ions to migrate to the nearest equivalent interstice.

The chance that so extensive articulated assemblies carry from the

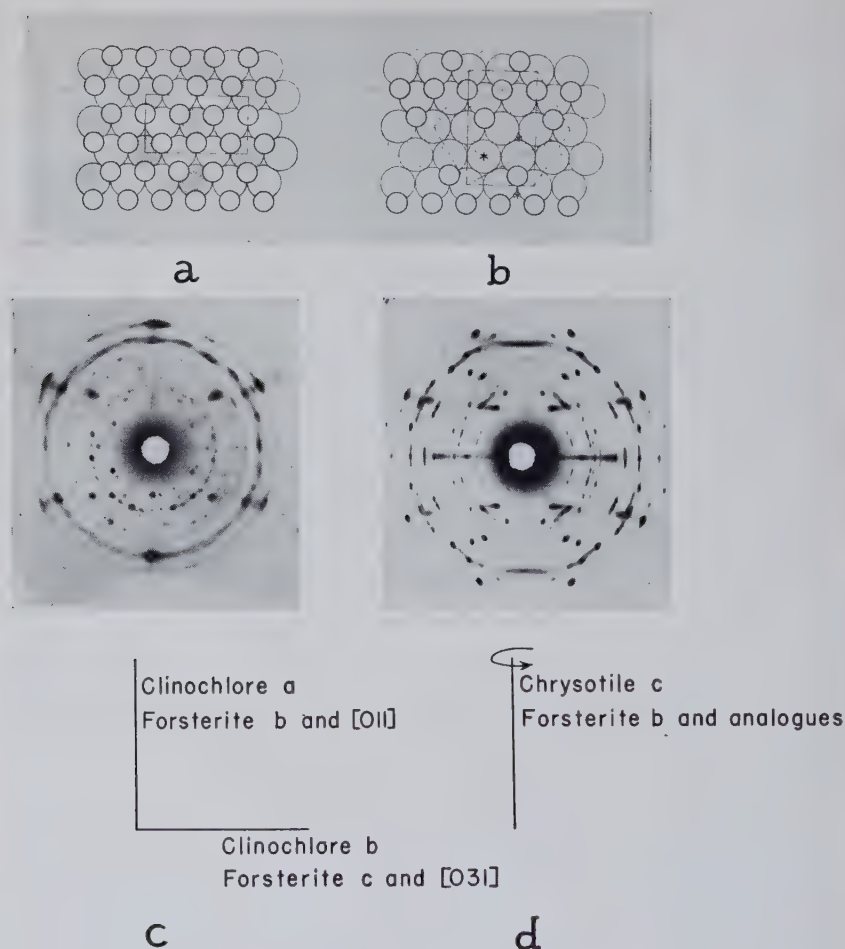


FIG. 2. The relationship of corresponding octahedrally coordinated levels in (a) clinochlore or chrysotile and in (b) forsterite. The diffraction diagram (c) represents a view along c for clinochlore or a for the forsterite growths. The diagram (d) is the synthetic fibrous forsterite product. In each case sufficient diffraction features of the respective unreacted starting materials show in the diagrams to mark the relative orientation before and after firing.

one structure to the other by other than lineal descent is remote. It seems therefore to be inferred that the stability of the brucite arrangement of octahedra is such that in the expulsion of water from its derivatives, it is the silica rather than the magnesia coordination which is disrupted. This is an apparently significant difference between octaphyllite and heptaphyllite octahedral arrangements.

THE SYNTHESIS OF QUARTZ

The synthesis of quartz from a crystalline silicate under the conditions of rapid temperature rise maintained in analysis is a matter of some concern. The quartz phase not only develops suddenly at temperatures of the order of 1000°C , the development being correlative with a sharp exothermal effect, but the crystallites are even subject to moderate grain growth in soaking periods of a few hours, before eventually transforming into cristobalite. The further complication that the quartz so synthesized on cooling to room temperature inverts in some cases and in others does not is probably less a surprise than is the synthesis itself.

The original concept of the layer silicates was drawn from observations of the dimensional compatibility of brucite and the cristobalite-tridymite type of tetrahedral layer. Probably one should think that, if a quick crystallization of silica were to occur at all, the modification should be one of those forms, but that does not seem to be the case.

Being granted that a structural inheritance can be traced from an old to a new phase in a particular instance, it seems reasonable to test whether any inheritance can be postulated that might govern the transition from linked layers to quartz in this instance.

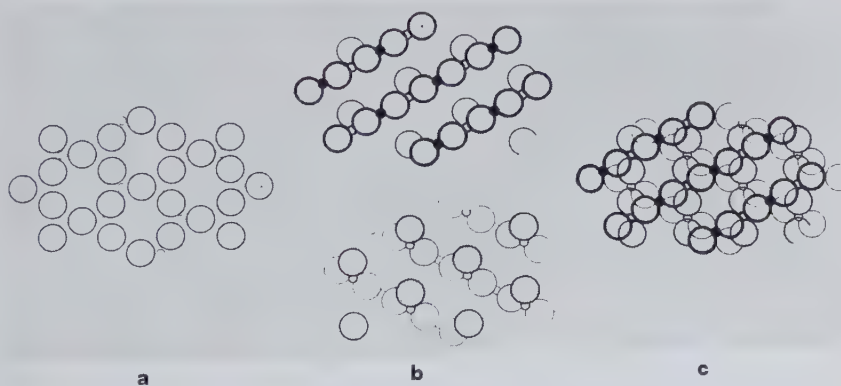


FIG. 3. The articulation of tetrahedrally arranged montmorillonite surfaces to form quartz.

- a. A montmorillonite surface.
- b. Two distorted arrangements of a.
- c. A superposition of the two levels b.

In Fig. 3 are sketched two sets of linked tetrahedra, representing adjacent montmorillonite surfaces, in which oxygens are partitioned into two kinds. Suitable articulation of these pairs actually does simulate the quartz arrangement of height equivalent to one complete unit cell and

the new grouping effects a localized gain in density of the order of about 10%.

This postulation of the articulation of montmorillonite surfaces into the quartz arrangement is a consequence of extended clean clay mineral surfaces. To articulate two such surfaces into a segment of the cristobalite structure would require the introduction of additional oxygen between surfaces to complete the coordination of any silicons which might provide valence bonds between layers.

Acid activated clays retain considerable quantities of $\text{SO}_4^{=}$, presumably on layer surfaces. These materials fire directly to cristobalite with a prominent exothermal effect around 1000°C . It is presumed that the presence of these interlayer sources of oxygen can condition the direct, energy-releasing articulation into cristobalite for such materials, even though in their natural condition they would fire to quartz.

MULLITE FROM PYROPHYLLITE

The synthesis of mullite from pyrophyllite appears to offer an example intermediate in facility of the synthesis and in clarity of the structural inheritance. Through the temperature range in which mullite is demonstrated to form there is no prominent exothermal feature, but only a broad ill-defined effect. There is again, however, a clear predisposition in the orientation of the synthesized mullite needles from which a structural relation may be deduced.

Figures 4*a* and *b* are illustrative diagrams. The (*a*) diagram is the monochromatic pinhole diffraction effect from a subparallel splinter aggregate of a fibrous pyrophyllite. Axial relations for the aggregate are sketched below. The (*b*) diagram is the resultant fired product. It consists of three superposed rotation diagrams for mullite, as rotated about *c*. Each diagram is 120° from the other two, and the three mullite *c* axis traces are parallel with the original pyrophyllite poles [020], [110], and [110].

The structural implications are reminiscent of those deduced for the forsterite synthesis. In the octahedrally packed layers in pyrophyllite, the orientation of each octahedron with respect to *b* and its analogues is precisely that of the orientation of each alumina octahedron with respect to *c* in the coordinated strings which extend along the center and corners of the mullite cell. In the absence of an actual analysis of the configuration in the octahedrally coordinated portion of the anhydride of pyrophyllite, one cannot follow the precise redistribution of aluminum ions among the apparently undisturbed octahedra. It does seem justifiable to conclude, though, that the octahedra are undisturbed and that the building of mullite strings, in this case without sharp exo-

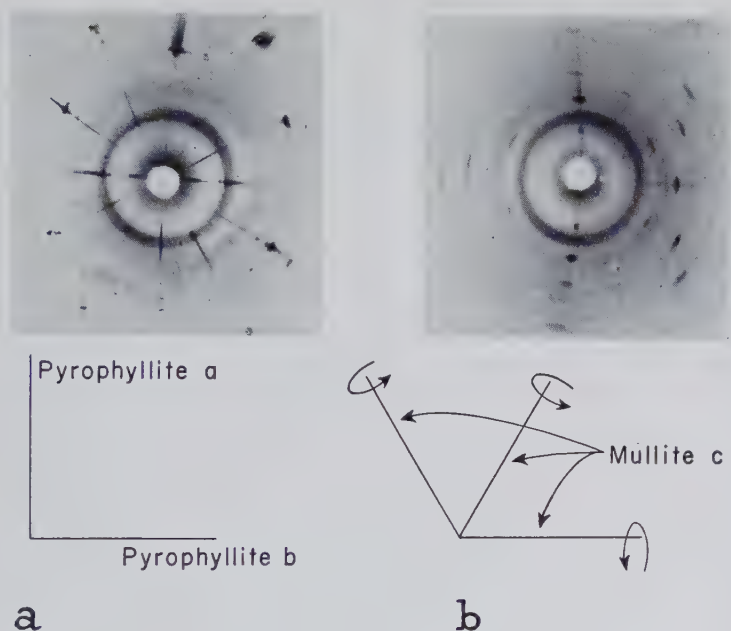


FIG. 4. Pyrophyllite and mullite.

a. Subparallel splinter aggregate of pyrophyllite. The three oxygen ion chain directions of the octahedral layer are horizontal and at the two 120° rotation positions in the plane of the film.

b. The mullite developed by firing splinter *a*. The three *c*-axis rotation diagrams demonstrate that all *c*-axes lie in and only in the three chain directions of *a*.

thermal effect, is merely less readily accomplished than the building of the same chains in kaolinite, which does afford a sharp exothermal effect.

THE THERMAL CURVES

The latitude of variation in the actual thermal analysis curves observed for the number of different minerals to be considered is such that they have not been set in order with respect to any particular generality.

The several groups of curves are simply reproduced in Figs. 5–10 without individual comment. In supplement a skeletal tabulation of respective constitutions at pertinent points in the firing sequence is arranged as a synopsis of the data in Table 3.

DISCUSSION

On a number of occasions, thermal curves of this sort have been reproduced for which it has been clearly established that prominent endothermal effects in two ranges around $100\text{--}250^\circ\text{C}$ and around $400\text{--}700^\circ\text{C}$

TABLE 3. GENERAL SUMMARY OF FIRING PRODUCTS FROM VARIOUS CLAYS

High alumina	900° C	1000° C	1100° C	1200° C	1300° C
Kaolinite		mullite (1)		mullite (1) cristobalite (2)	
Endellite		mullite (1)		mullite (1) cristobalite (2)	
Diaspore	α -Al ₂ O ₃ (1)				
Gibbsite	γ -Al ₂ O ₃ (1)	α -Al ₂ O ₃ (1)			
Bauxite (Kaolinite & gibbsite)					
Montmorillonite group					
Otay, Calif.		β -quartz (1) enstatite (3)	cristobalite (1) β -quartz (1) enstatite (2)	cristobalite (1) cordierite (1)	cristobalite (3) cordierite (1) periclase (3)
Tatatila, Vera Cruz		β -quartz (2)			cristobalite (1) mullite (2) cordierite (2)
Upton, Wyo.	spinel (1)	spinel (1) α -quartz (2)	spinel (2) cristobalite (1)		mullite (2)
Cheto		β -quartz (1) anorthite (?) (3)	β -quartz (1) cristobalite (3) anorthite (?) (3)		cristobalite (1) cordierite (1)
Palmer, Ark.		spinel (1)	spinel (1) quartz (3)	cristobalite (1) spinel (1) mullite (1)	
Sierra de Guadalupe		spinel (1)	cristobalite (1) spinel (1)	cristobalite (1) spinel (1) cordierite (2)	cristobalite (1) cordierite (1)
Harris Co., Texas		spinel (2) cristobalite (3)	cristobalite (1) spinel (1)	cristobalite (1) spinel (1) mullite (2)	cristobalite (1) mullite (2)
Pontotoc Co., Miss.		spinel (1) α -quartz (2)	cristobalite (1) spinel (1)		cristobalite (2) cordierite (2)
Beidell, Colo.					mullite cristobalite
Fairview, Utah				mullite (1) cristobalite (1)	mullite (1)
Wagon Wheel Gap	spinel (2)	cristobalite (1) mullite (3) spinel (2)			cristobalite
Woody Nontronite					mullite cristobalite spinel

Parenthetic numbers signify: (1) important, (2) moderate, and (3) minor.

are related respectively to loss of molecular and hydroxyl water. In the present discussion, attention will be confined to reactions which follow the second water loss.

The three-layer anhydride phase, present between the 400–700° reaction and the subsequent endothermal effect about 200° higher, is not



FIG. 5. Miscellaneous hydrous minerals.

- A. Kaolinite, Dry Branch, Georgia.
- B. Endellite, Eureka, Utah.
- C. Diaspore, Chester, Massachusetts.
- D. Gibbsite, Saline County, Arkansas.
- E. Bauxite (Gibbsite and Kaolinite), Irvington, Georgia.

strictly anhydrous. Additional water losses of the order of one-half to one per cent are readily demonstrated in weight vs. temperature curves, but the magnitude of the loss is too small to hope that its significance might ever be deduced from diffraction data. Nor does it seem justified in the absence of an instantaneous weight record to speculate on whether the amount of water loss under analysis conditions is commensurate with the indicated energy effect.

Beyond the temperature of water losses, over-all compositions of the separate systems are not subject to change (ignoring instances of reducible iron oxide content). The problem is thus not to account for the simplicity of a curve, but to account for its complexity. There seems to be no reason to expect that a system would attempt to approach equilibrium by successive reactions. It is here that the role of structural inheritance becomes apparent. Predisposition to crystallize as a phase which "doesn't belong" affords starting material for a subsequent reaction, with a possible subsequent energy effect.

Probably in no case does the intensity of an inheritance energy effect or the estimated abundance of that phase in the diffraction diagram bespeak the entire content of that chemical component. In general, though, it appears to be true that great abundance of such a phase correlates with greater intensity for subsequent effects in the same system. For those curves which show two prominent exothermal effects in the high temperature range then, it is indicated that the first is a consequence of the coordination scheme of the natural mineral and the second is a consequence of its chemical composition.

Perusal of the various three-layer clay mineral curves brings out that separate thermal traces tend to differentiate themselves into two separate types, those which present an *S*-shaped feature (as Fig. 6*c*, 7*a*, 7*c*, 8*d*), and those which exhibit a distinct shoulder (as Fig. 6*a*, 6*b*, 6*d*). Each presumably represents an energy absorption followed by energy evolution, the two effects being simply unresolved for the former case. In terms of phase development, however, the separate types are clearly differentiated. Clays of the *S*-shaped curves contain proportions of spinel type oxides, following this reaction, which are proportional to the observed magnitude of the thermal effect. The second group, in which crystallization seems to be somewhat delayed, are those in which the first prominent new phase to appear is quartz. The new phase is thus seen to develop in the one case from the octahedral region in the clay, and in the other case from the tetrahedral. It is perhaps to be noted that the trace depicting crystallization of forsterite (based on the octahedral portion) from chlorite is an exaggeration of the *S*-shaped type.

In the comprehensive tabulation of montmorillonite group analyses included in the recent survey by Ross and Hendricks (9) corresponding

specimens in the two studies fire to quartz if they appear in Ross and Hendricks table with Si^{++} population near 4.00, and fire first to spinel if there is substantial substitution of Al^{+++} in tetrahedral coordination. It

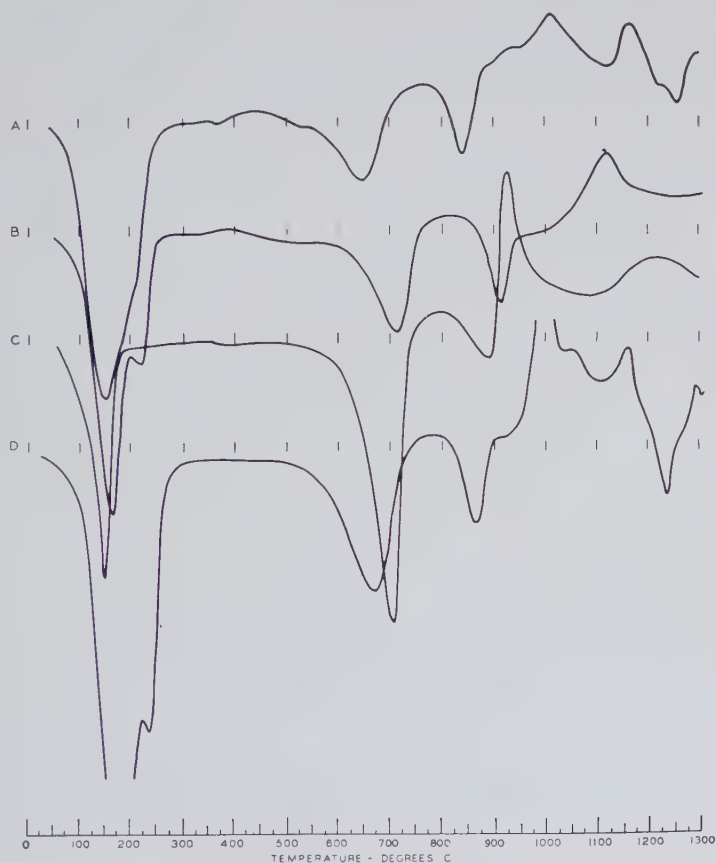


FIG. 6. Montmorillonite.

- A. Otay, California.
- B. Tatatila, Vera Cruz, Mexico, U. S. Nat. Mus. 101,836.
- C. Upton, Wyoming.
- D. Chambers, Arizona.

is inferred that regularity in the composition of the tetrahedral scheme endows it with ability to maintain its unity under conditions which disorganize substituted layers.

Clays of either type, which have undergone either of the foregoing syntheses, may show an additional fairly prominent reaction near 1200°C . The new phase encountered here seems to be either mullite or cordierite with about equal frequency. One reaction having already taken place

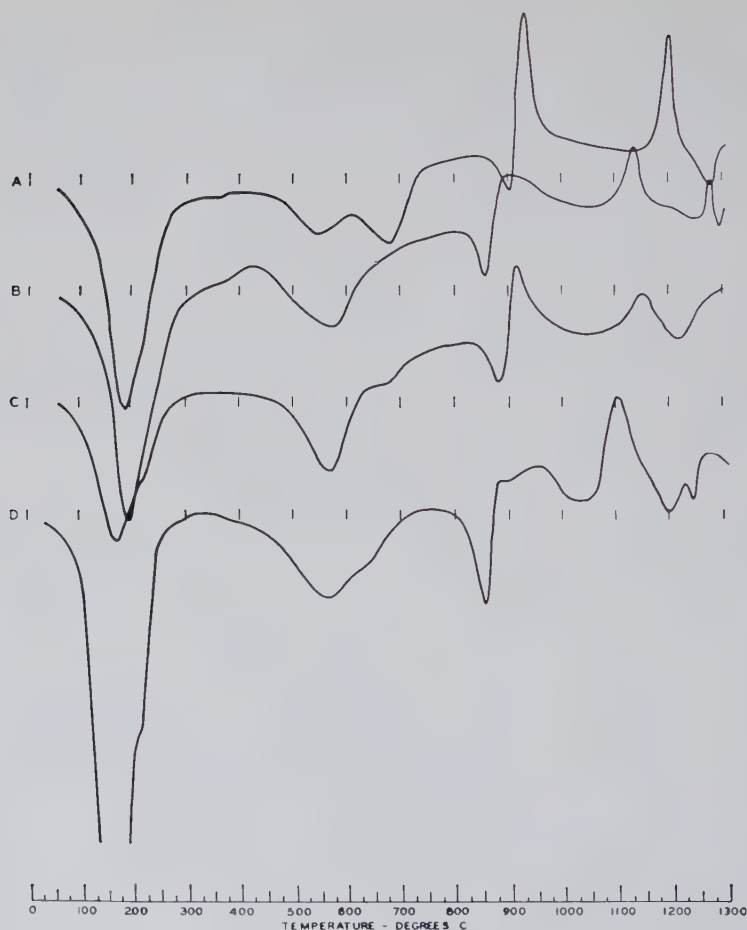


FIG. 7. Montmorillonite Group Clays.

- A. Palmer, Arkansas.
- B. Sierra de Guadalupe, Atzoopozalco, Mexico, U. S. Nat. Mus. 7591.
- C. Harris County, Texas.
- D. Pontotoc County, Mississippi.

there is no reason to believe that either of these crystallizations are inheriting characteristics from the starting material. They seem to be predetermined only by the chemical composition of the system, and have in common only the properties of density and hardness which would indicate that both would evolve heat in formation from the respective raw materials present.

The spinel forming specimens all develop more or less cristobalite around 1000° or 1100° C., without going through the quartz inter-

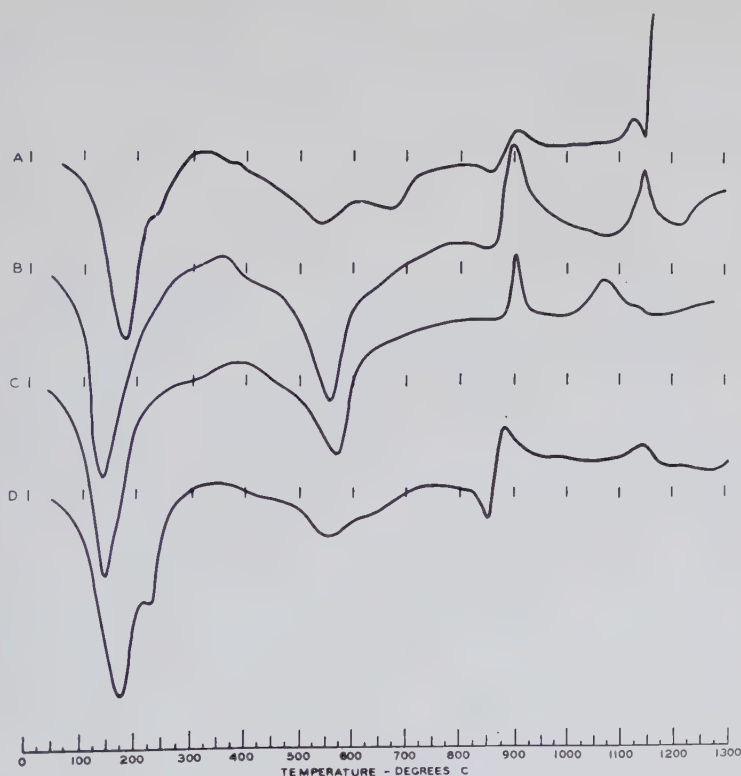


FIG. 8. Materials Often Called Beidellite.

A. Beidell, Colorado, U. S. Nat. Mus. 93239.

B. Fairview, Utah.

C. Wagon Wheel Gap, Colorado, U. S. Nat. Mus. 94963.

D. Nontronite, Howard County, Arkansas.

mediate stage. No separate feature appears in the curves to indicate any energy effect associated with the growth. Since this is true also of opal fired alone, it is assumed that any exchange involved is not picked up by the method. Cristobalite is actually effectively somewhat less dense than the condensed layer configuration, and might even require an energy input to crystallize under the analysis conditions.

SUMMARY

1. Endothermal effects up to 900–1000° C are normally occasioned by the expulsion of some volatile constituent.

2. Exothermal effects of moderate magnitude may, when they immediately follow endothermal effects, be evidenced merely in the more abrupt return of the test specimen to reference temperature.

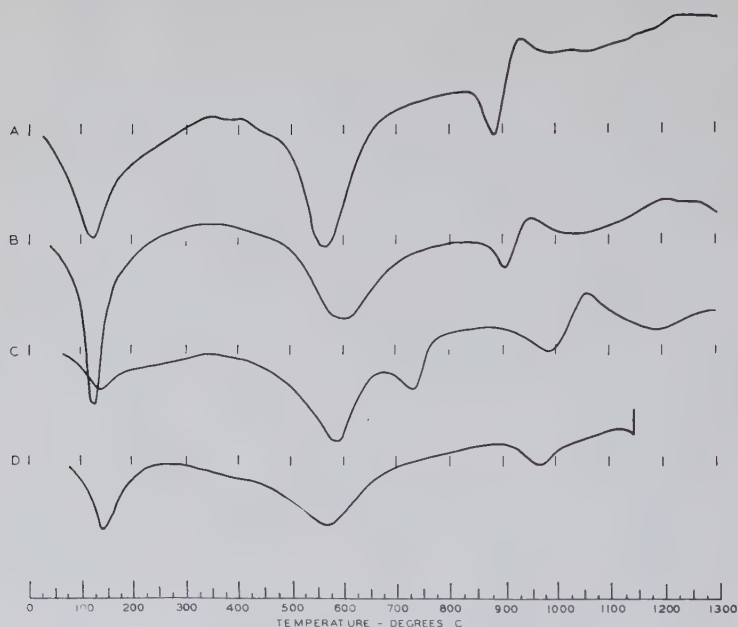


FIG. 9. Common Potassium-Bearing Clays.

- A. Vermilion County, Illinois.
- B. Grundy County, Illinois.
- C. Sarospatak, Hungary ("Glimmerton" from U. Hofmann, Rostock).
- D. New Jersey green sand, University of Illinois collections.

3. Sudden sharp exothermal effects are occasioned by the incorporation into new phases of large articulated units from reactant structures without catastrophic rearrangements within the units.

4. Secondary exothermal effects may be observed for crystallization of phases whose development is conditioned only by the chemical composition of the system.

NOTE

Since the preparation of this article, Roy and Osborn (10) have stabilized high quartz at room temperature with lithium, and Brindley and Ali (11) have analyzed the mechanism of forsterite growth in fired chlorite flakes.

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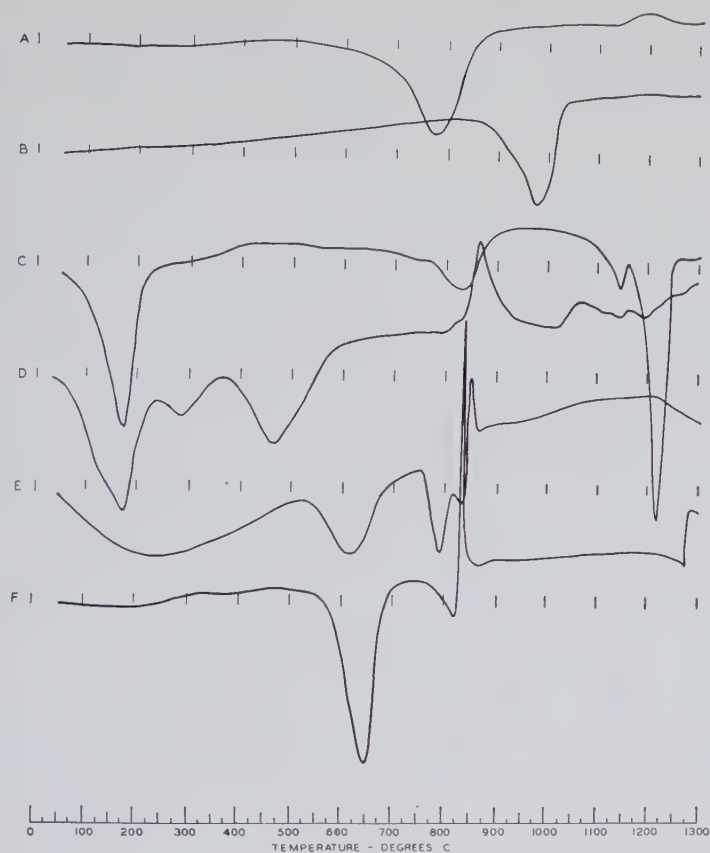


FIG. 10. Miscellaneous Related Minerals.

- A. Pyrophyllite, North Carolina.
- B. Talc, Vermont.
- C. Hectorite, Hector, California.*
- D. Attapulgit, Quincy, Florida.
- E. Prochlorite, Chester, Vermont.
- F. Clinocllore, Brewster, New York.

* The unique feature near 1200° is the inversion to clino-enstatite under the mineralizing influence of fluorine in the natural clay.

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FRACTOGRAPHY AS A MINERALOGICAL TECHNIQUE*

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SUMMARY

Fractography, the special microscope technique developed during the past decade in the field of metallurgy, is applied to a preliminary study of several minerals for purposes of introducing fractography to mineralogists as a possibly useful petrographic method.

First, a general classification of fracture patterns is established to separate the markings relating to structures intrinsic to the crystal (Type I) from those caused by local resolutions of the transient fracturing stress (Type II). Examples are shown using metals and synthetic chemical crystals; and these in turn provide a further subclassification of the Type I patterns—which are the patterns of greatest significance—into (1) Type Ia, representing the crystallographic directionalism of the lattice structure, (2) Type Ib, expressing crystal imperfection following from failure to attain an ideal lattice structure, and (3) Type Ic, disclosing the presence of extraneous phases.

On the basis of these categories, a number of minerals are studied fractographically, including plagioclase, orthoclase, muscovite, fluorite, dolomite, twinned calcite, and optically pure calcite. The patterns are found to have two aspects of broad importance: (1) petrographic and (2) micellar.

With regard to the first, the following phenomena are illustrated: (a) twinning, (b) parting on twin planes, (c) perfect cleavage, (d) intersecting primary and secondary cleavages, (e) fine superficial striae, possibly related to twinning, (f) occluded interlamellar phases in muscovite, (g) inclusions, both solid and fluid, and (h) markings of unknown origin.

As for the micellar aspect, reference is made to the current widespread discussions of theories attempting to reconcile the vast differences between “ideal” and “real” crystals, and specifically to the recent “micellar theory” which proposes a universal subdivisional nature for the solid state inherited from a micellar condition in the liquid. Fractographs of the minerals, as of the metals, prove to be rich in evidence for such a mosaic or micellar constitution; and some elaborate imperfection patterns are presented.

INTRODUCTION

During the past decade there has been developed in the field of metallurgy a specialized micrographic technique now known as “fractography” (1, 2, 3). As the name implies, this technique concerns the study of detail on fracture surfaces. Because mineralogy has concerned itself with the appearance of fractures virtually since that science began, one might wonder whether there is anything further to be gained from it.

In the field of metallurgy, the situation is somewhat different, for the fractures of most metals have a roughness and a multiplicity of minute crystals which make direct microscopic observation difficult and forbidding. De Réaumur (4) made some historically interesting sketches of

* From research conducted in the Laboratory of the senior author in Baltimore, Md., under sponsorship of the Office of Naval Research.

** Since Aug. 1, 1950, with Horace T. Potts Co., Philadelphia, Pa.

*** Geologist, Brainerd, Minnesota. Deceased August 28, 1950.

the fractures of iron and steel in 1722, based upon microscopic examination which probably followed contemporary mineralogical practice; nevertheless, after Martens' work (5) in 1887 was abandoned in favor of the method of direct study of fractures, what has now become conventional metallography—the cutting, polishing, and etching of solid sections and their study by reflected light.

Glancing briefly at the parallel history of mineralogy (6), one finds Henry Clifton Sorby in 1849 publishing in his "Calcareous Grit of Scarborough" the first work on thin sections of minerals. His attention had been attracted to a study by Williamson on thin slides of fossil wood. Sorby wrote again on "Slaty Cleavage" in 1851; and in 1857 he presented a paper on "The Microscopic Structure of Crystals" before the Geological Society of London. For many years Sorby's work received little attention, although today the thin-section technique is the rampart of petrography.

An attempt by Sorby to apply the thin-section technique to metals failed, of course; but this led to his development of the polish-etch method of modern metallography. The two fields of mineralogy and metallurgy thereafter separated, so far as techniques for discerning structure and constitution were concerned.

In the development of fractography, however, attention has again been turned toward the detail to be found on cleavage and fracture surfaces. Certain technical improvements in microscope construction have appeared since Martens' time; and manipulations have recently been found which enable the direct observation of surfaces, almost regardless of their roughness. Furthermore, the technique as it is most fruitfully applied concerns high magnifications almost exclusively—from one- or two-hundred diameters up to the limits of the lens system. This change in scale of observation carries the work into categories not touched by the early workers.

Consequently, the aim of the present writing is to present to mineralogists a brief and generalized treatment of various exemplary metal and mineral systems for purposes of calling the attention of that profession to possible contributions which might be made by fractography applied to minerals. Some of the work to be shown has undoubtedly been observed before by certain mineralogists, and probably more effectively. Nevertheless, some of its aspects are new.

In addition, because of the rapidly mounting interest in theories for subtle substructures in the solid state—the so-called mosaic (7) and micellar (8)* theories, for example—a review of details found on nascent fracture surfaces in all solids is well warranted. As this paper will clearly

* See Appendix for an outline of the micellar theory.

illustrate, fractography discloses impressive observations of a subdivisional architecture within the crystal, observations which can have much to do with the formulation of an understanding for certain characteristics of solids which at present have no satisfactory explanation. One is here reminded of Haüy, the discoverer of the Law of Rational Indices and the founder of modern crystallography, who a century and a half ago described crystals as physical composites of a "crystal molecule" (9) and sketched the gross structures as a brickwork of submicroscopic perfect crystallites. His concept was driven under by the impetus of atomic theory, the space lattice, and particularly *x*-ray diffraction; nevertheless, there has redeveloped in the past quarter century some rapidly mounting evidence that such subdivisional structure does exist between the molecule or unit cell and the single crystal.

This text will accordingly divide its attention between: (1) evidences in fractographs having promise from a petrographic or crystallographic standpoint, and (2) evidences for subdivisional architecture having significance for theories of the solid state.

EXPERIMENTAL METHOD

A number of papers have already appeared on fractography; and to avoid some repetition here, Ref. No. 3 has been listed in the bibliography. In that paper a complete bibliography of fractographic studies through 1948 can be found, and these in turn can be used to supply details of the technique as developed to that date. The first three papers listed in the present bibliography also contain descriptions of the two types of fractographic stages used thus far. These descriptions are not necessary to repeat here because the fractographic stage is little other than a simplified orienting mechanism, whose general principles are well known to mineralogists.

In brief outline, an orienting mechanism holds a fractured chip in a clamp or in a plasticene cup, with the fracture facet roughly perpendicular to the microscope axis. A lens providing a magnification in the order of 100 \times is brought toward the fracture face until a rough focus is obtained. The stage is then rotated and shifted until some portion of the fracture field attains good perpendicularity and yields a usefully flat field. Such an area will announce its arrival in the field by a flash of light. A stronger lens can then be inserted, if desired, the focus refined, and the field explored.

Magnifications up to the limit of the microscope are readily used, at least on opaque bodies; and magnifications greater than 100 \times are generally recommended.

Illumination by any of the common processes is applicable, including:

(a) vertical illumination, (b) oblique illumination, (c) dark-field, (d) phase-contrast, (e) sensitive tint, and (f) polarized light; however, the procedure for the work as developed to date, and as used in this paper, comprises vertical illumination (inverted metallurgical microscope) *slightly obliqued to provide surface contrast*. The importance of obliquing, particularly for photography, cannot be overemphasized.

Specimens include any solid whose fracture surface can be isolated for study. If promontories interfere, they can be removed by delicate hand grinding operations; but often the promontories themselves have on them informative structures. While the facet can be polished or etched, or treated much as desired, the primary basis for fractography lies in the nascent, unetched, and untouched fracture surface.

PATTERN TYPES

Before considering the fracture patterns, two broad types of markings must be defined:

Type I: Patterns developed by factors intrinsic to the solid.

Type II: Patterns imposed by factors extrinsic to the solid.

In the first, Type I, the path of fracture follows a traverse influenced by, and hence expressing, the structural forms and directional weaknesses within the solid itself, and therefore reveals directly the architecture of the specimen. This classification, Type I, contains by far the more useful information; and because of its prolific registrations, a number of subclasses can subsequently be named.

For example, in Fig. 1 there are reproduced two fractographs taken of cleavage surfaces in cast bismuth metal. The upper, representing cleavage on the basal (0001) plane, discloses two broad bands which are shaded gray by the oblique illumination and are known to be twins on $\{10\bar{1}4\}$. Also, on close scrutiny of this fractograph one will perhaps discern, even in the reproduction, faint parallel lines of "striae" running along three sets of directions at 60° to one another. While the origin of these has not been positively identified (10, 11, 12), the point is readily allowed that the two groups of phenomena—twins and striae—have created a pattern on the fracture traverse because of a fundamental crystallographic directionalism. *Type Ia* can therefore be named as a subclass referring to *deflections in the fracture traverse caused by the directionalism of the atomic lattice structure*.

In the lower fractograph of Fig. 1, cleavage on an inclined plane of $\{10\bar{1}1\}$ form is depicted. Contrast to the basal cleavage pattern is marked. Registrations of the twin bands again appear, but much altered in structure and, of course, in respective angularities, since the planar intersection is now $\{10\bar{1}4\}$ upon $(10\bar{1}1)$. One can orient this pattern by recogniz-

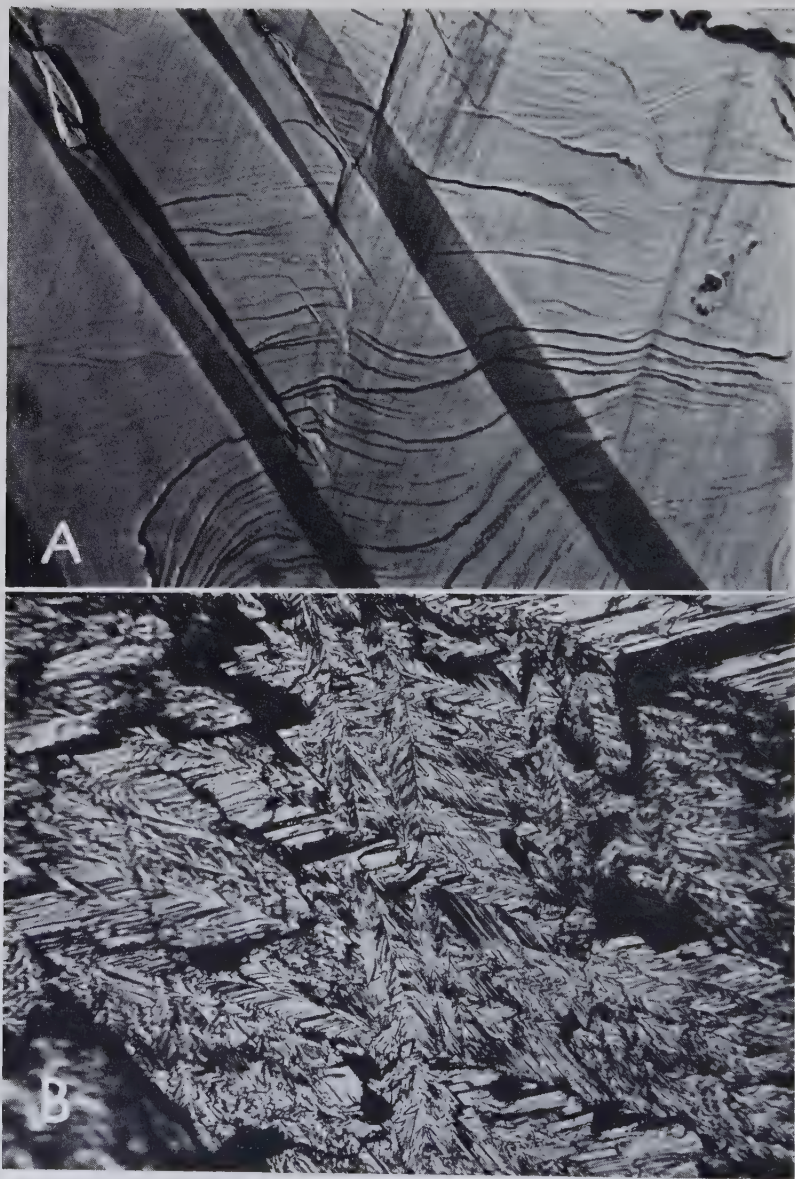


FIG. 1. Fractographs of cleavage in commercially pure cast bismuth illustrating Types Ia and Ib patterns for a homogeneous constitution. (Above) Basal cleavage on (0001) at 550 \times . (Below) Secondary cleavage on (10 $\bar{1}$ 1) at 170 \times .

ing the small sharp profiles as (0001) cleavages intersecting the (10 $\bar{1}1$) plane of the fractograph. The herringbone pattern then displays the twin by the small laminar basal cleavages now reoriented along the twinned layer.

Since it is not the purpose of the present writing to describe the crystallographic details of deformation and fracture in bismuth, discussion of this second fractograph will turn its attention instead toward the remarkable substructural detail. Here is direct evidence of an impressive kind that this single crystal of bismuth (the grain boundary lies far outside the photograph) comprises an elaborate substructure of a mosaic or micellar type (8), showing here particularly as laminae. Because this observation will become much fortified by later evidence, the feature can be at least tentatively accepted as warranting a *Type Ib* subclass for patterns expressing *directionalism resulting from imperfection in the development of the ideal lattice structure*. This will include in turn a broad category extending from the postulated micellar structure up through such disturbances as dendrites and grain boundaries. One will observe, however, that both subclasses Ia and Ib refer specifically to physical nonuniformities within a single homogeneous phase. These are structural features which can concern the presence of a second phase, but do not require it.

On the other hand, turning to Fig. 2, one finds strongly marked fracture characteristics which are again clearly intrinsic to the crystal, but are specific registrations of a second phase. The specimens here are molybdenum metal, vacuum-cast by an electric arc process (13, 14). In the upper fractograph there appear featherlike forms, now known to represent molybdenum carbide; and their position is characteristically intergranular.

In the lower fractograph, similar "carbide feathers" are again evident in the upper portion of the field; and encroaching upon this field are dendritic growths of molybdenum oxide, also intergranular. The chemical reaction of these two compounds under the high-temperature conditions of melting yields escapable carbon oxide gas; and one can observe the disappearance of the carbide toward the lower portions of the field. These two fractographs display the phenomena, incidentally, which control the forgeability of this metal.

Molybdenum displaying the upper fracture pattern is forgeable. That displaying oxide is nonforgeable and must be remelted.

Consequently, the fractographs in Fig. 2 clearly define a third subclassification under Type I. The pattern is intrinsic to the system, but *refers to the intervention of a heterogeneous constitution* and will be defined as *Type Ic*.

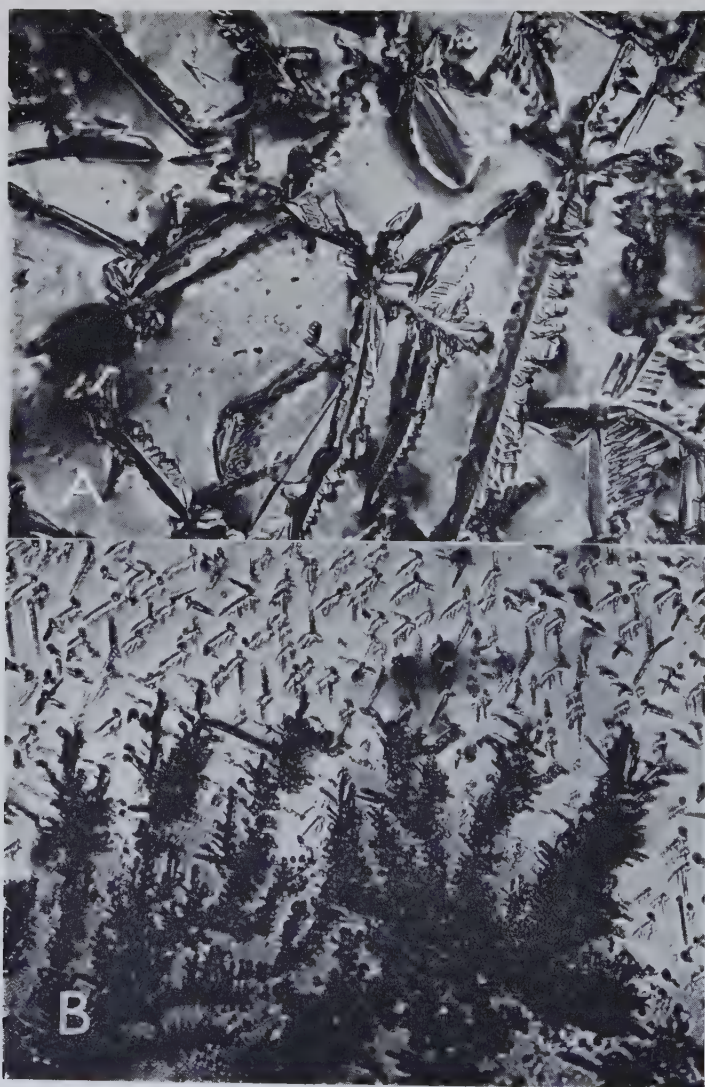


FIG. 2. Fractographs of cast molybdenum metal illustrating Type Ic patterns for heterogeneous constitution. (Above) Featherlike intergranular structures expressing carbide. 440X. (Below) Dendritic growths of intergranular oxide encroaching upon featherlike carbides as shown above. 320X.

Proceeding now to Fig. 3, one observes a pattern which is distinctive both in appearance and in cause. This is the Type II classification, resulting from factors extrinsic to the specimen—specifically stress. While subclassifications can be added to this category likewise, the present discussion will limit itself to a broad consideration of stress as the extrinsic factor.

Thus, the upper fractograph illustrates the typical pattern relating to the traverse of fracture through a matrix which imposes none of its own directionalism upon that traverse. The pattern is essentially a stress pattern, revealing the undulations and nodes of the shattering wave; and the path is similar for vitreous or amorphous bodies and for crystals exhibiting minor directionalism. Here the specimen is a synthetic crystal of ammonium dihydrogen phosphate, whose tetragonal structure perhaps should be expected to show Type I effects. In fact, it does to some extent, as is shown elsewhere (15); but there also occurs much Type II traverse, of which Fig. 3A is an example.

In the lower fractograph of Fig. 3 another fracture of this material is shown, photographed with dark-field illumination. The pattern is again predominantly Type II, although an elaboration occurs which is somewhat suggestive of lamellar Type Ib effects.

In summary, these three sets of fractographs convey the principal generalizations to be understood in interpreting the following fractographs of minerals. All of the Type I subclasses are generic to the study of the minerals to be discussed. Type II patterns are of no particular interest for the work at hand, and are to be distinguished only for purposes of their separation from the more informative markings.

In closing the discussion of pattern types, a particularly interesting example of the fractographic technique is given in Fig. 4. The specimen is isometric alpha iron (ferrite) which has been rendered brittle by absorption of hydrogen gas during treatment in sulfuric acid. The sample is polycrystalline, typical of irons and steels, whose grains are much smaller than those crystals commonly explored in mineralogy, and which therefore provide a problem for which fractography is specifically designed. The fracture of the specimen presented a typical rough appearance; but a single cleavage facet was selected and photographed. The corresponding half of the specimen was next oriented on the stage and explored until the matching facet was found. It, too, was photographed, at the same orientation and magnification. The pair of fractographs in Fig. 4 then provide an "obverse" and "reverse" view of the fracture traverse through this particular grain.

In these two matching patterns one will first note the rectilinearity, which immediately identifies it as being predominantly Type I. The

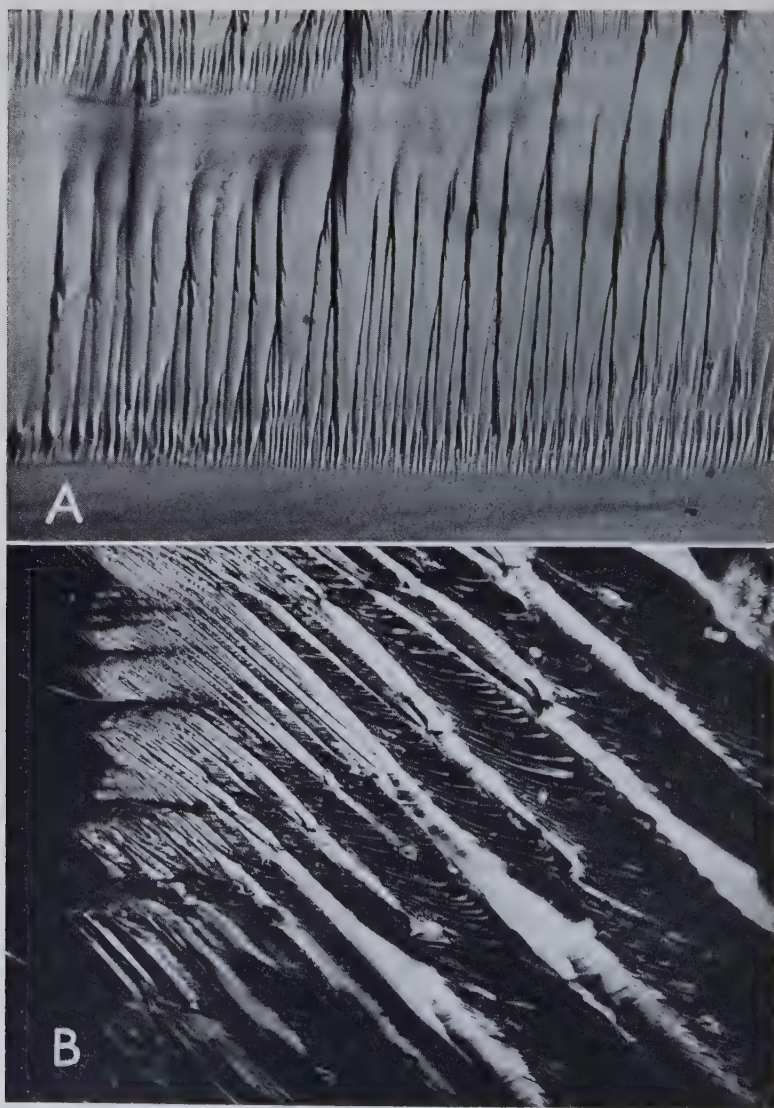


FIG. 3. Fractographs illustrating Type II patterns in synthetic ammonium dihydrogen phosphate crystals. (Above) Typical pattern caused by stress in a matrix exhibiting no directionalism. 100 \times . (Below) Type II pattern photographed with dark-field illumination and exhibiting fine-scale detail. 70 \times .

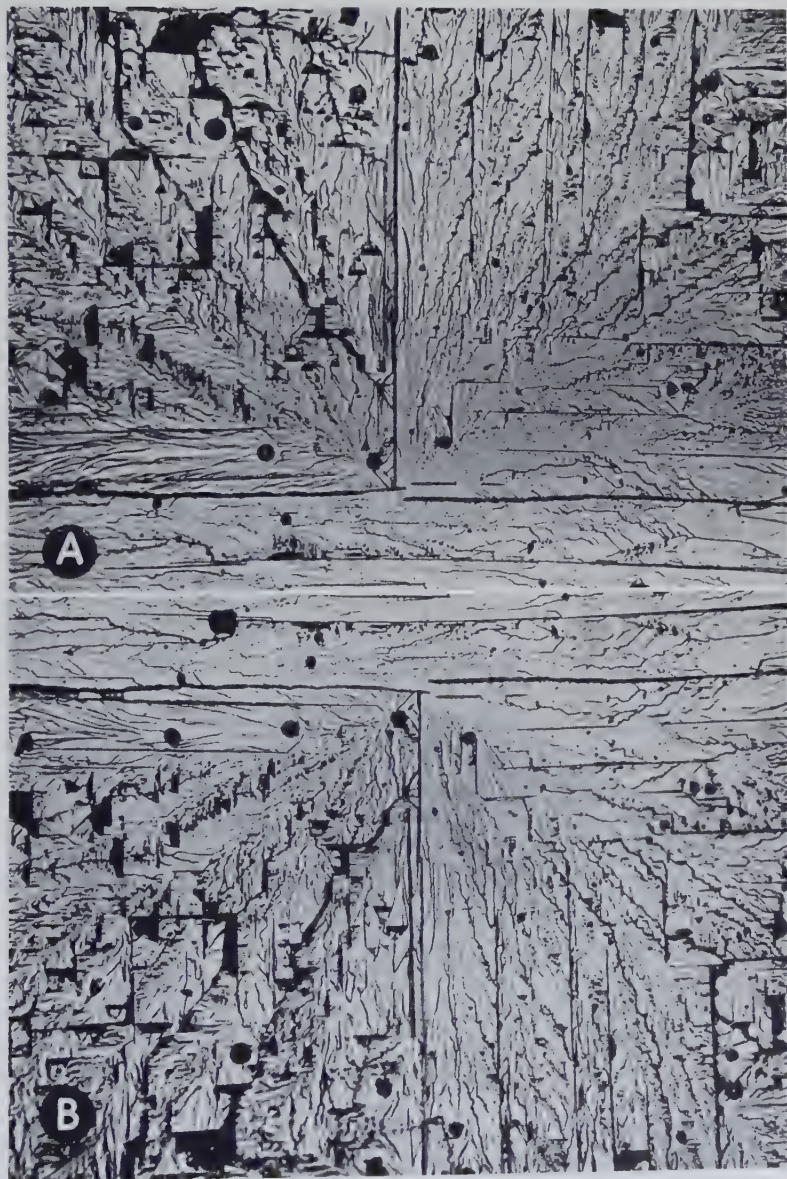


FIG. 4. Observe and reverse fractographs of a cleavage facet for a single grain within a polycrystalline specimen of isometric alpha iron made brittle by absorption of hydrogen gas. 235X.

face is (001), and the principal markings at 90° are intersecting (100) and (010) cleavages. Markings at other angles can usually be found on close observation to be stepwise composites of {100}, although some slip (gliding) activity on {110} or {112} is also possibly present. The subclass Type Ia identifies itself in the form of this crystallographic directionalism. Type Ib is strongly suggested in the minute blocklike formations, in conformity with the postulate of a mosaic or micellar structure (8), for cleavage would not suffer such perpendicular displacements if the path were not one of special weakness. Type IC can only be represented in this rather pure material, of course, by the pits and voids caused by inclusions of minute contaminating phases.

As for Type II, its patterns are virtually absent. The meandering or fanlike patterns could possibly be argued as a pattern of stress, but they can also be argued as an imperfection pattern referring to growth directionalism—the so-called “lineage” or dendrite pattern (16, 17). This distinction is often a subtle one; and the exact definition to be made between patterns from dendritic growth and those from stress remains to be determined. However, these are usually boundary cases of relatively minor importance. When they occur, experience with other systems where the origin of the pattern is quite certain will often allow a classification to be made. Where this cannot be done, the pattern will be so noted and left for subsequent study.

EXAMINATION OF MINERALS

In Table 1, the specimens used in this research are listed by name, crystal system, approximate composition, and source. In addition to these, others were also examined, such as olivine, talc, epsomite, thulite, magnesite, and quartz. Their description will not be included, principally for reasons of brevity, but also because of some photographic inferiority caused by such factors as poor reflectance and uninformative-ness of the patterns. The recorded work will suffice for the present purpose of introducing fractography as a possibly useful microscope technique and suggesting fields for subsequent special studies.

THE FELDSPARS

Plagioclase

Since one of the most important and most common of the mineral groups is that of the feldspars, plagioclase—the triclinic isomorphous series of Na-Al and Ca-Al silicates—is shown in Fig. 5. The specimen is labradorite; and the fractograph shows a system of parallel bands which can probably be accepted as albite twinning on the brachypinacoid {010}. The cleavage plane, which is the plane of the fractograph, can

TABLE 1. DESCRIPTION OF SPECIMENS

Material	Crystal System	Formula	Source*
1. Bismuth	Hexagonal-rhombohedral	Bi	Synthetic crystal (commercially pure)
2. Molybdenum	Isometric (BCC)	Mo	Climax Molybdenum Co. (vacuum cast)
3. Ammonium-dihydrogen phosphate	Tetragonal	$(\text{NH}_4)\text{H}_2\text{PO}_4$	Naval Research Lab. (synthetic crystal)
4. Iron (alpha ferrite)	Isometric (BCC)	Fe	Armco ingot iron
5. Plagioclase (Labradorite)	Triclinic	$(\text{Na}, \text{Ca})\text{Al} \begin{array}{ c} \text{Si} \\ \text{Al} \end{array} \text{O}_3$	Labrador
6. Orthoclase	Monoclinic	KAlSi_3O_8	Carbon Co., Mont.
7. Muscovite	Monoclinic	$\text{KAl}_3\text{Si}_3\text{O}_{10}(\text{OH})_2$	Okanogan Co., Wash.
8. Fluorite	Isometric	CaF_2	—————
9. Fluorite	Isometric	CaF_2	Ferry Co., Wash.
10. Dolomite	Hexagonal-rhombohedral	$\text{CaMg}(\text{CO}_3)_2$	Stevens Co., Wash.
11. Limestone	—————	—————	Snohomish Co., Wash.
12. Calcite	Hexagonal-rhombohedral	CaCO_3	(twinned)
13. Calcite	Hexagonal-rhombohedral	CaCO_3	(optically pure)

* See acknowledgment. Further details on the specimens of minerals can be obtained from G. M. Valentine and N. W. Buerger.

probably similarly be accepted as a basal $\{001\}$. These planes were not specifically identified here; but a method has been developed in fractography which allows this to be done (18). That is, photographs of two intersecting cleavages are mounted on an appropriate model, and the families of the active planes are then directly determined from the positions of the traces in the three dimensions. This technique has already

resulted in a corrected determination for certain deformation phenomena in bismuth and antimony (18).

In Fig. 6 two further fractographs of plagioclase are shown. Several features of interest appear in the upper photograph. First, there is a pair of thin parallel lines running diagonally from upper left to lower right.



FIG. 5. Gridwork of albite twins in labradorite. 135 \times .

which can be tentatively judged as twins because of (a) their similarity to twin markings in the previous figure, and (b) the fact that the meandering "tear lines" are not noticeably influenced by their presence, as they would be if the markings were cleavages. Second, there are three further geometric markings forming a spearhead toward the left of the field, none of which is parallel to the previous set. Each of these three lines deflects the intersecting tear lines, suggesting a development prior to fracture; and one of these—the horizontal marking—is visibly a cleavage, which almost perfectly bisects the angle formed by the other two. As for these remaining two markings, they seem outlined by pits

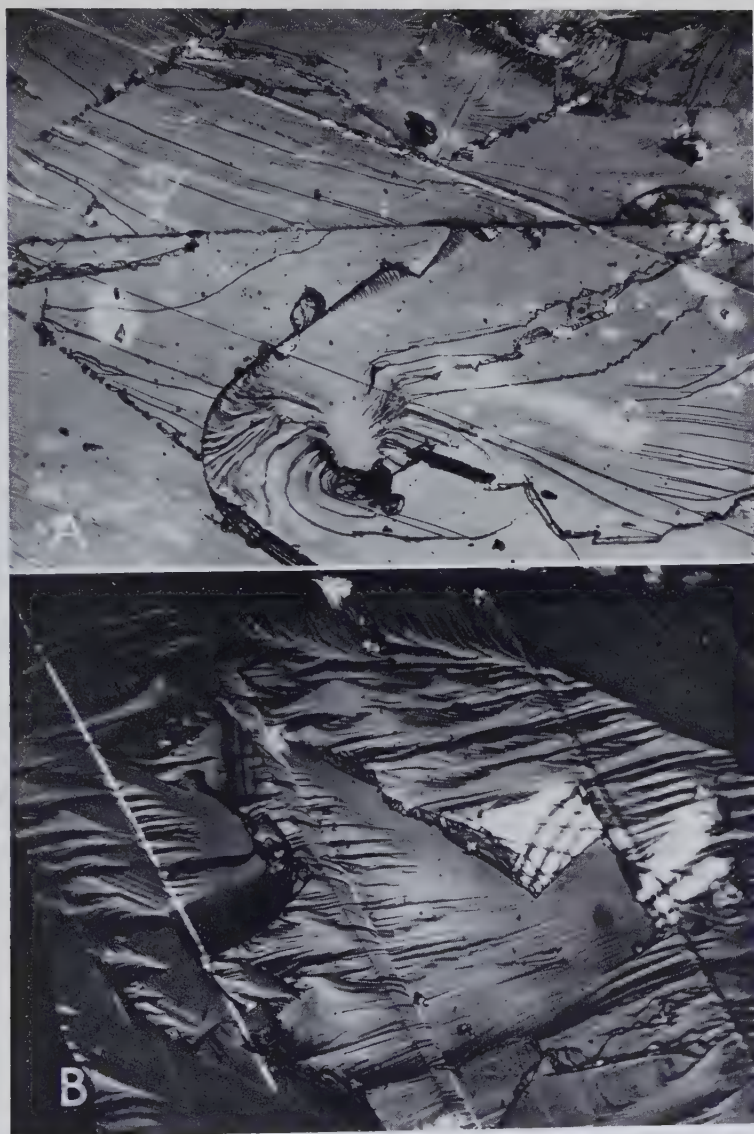


FIG. 6. Fractographs of labradorite. (Above) (a) albite twins, (b) intersecting cleavage, (c) parting on twin planes, (d) whorl pattern of tear lines nucleating at an inclusion, (e) fine-scale stepwise pattern in tear lines suggestive of mosaic structure and parting along minute twins, and (f) two intersecting pitmarked lines suggestive of a former growth face. 95 \times . (Below) Carefully shadowed surface detail showing (a) albite twins, and (b) fanlike patterns possibly relating to growth imperfection. 235 \times .

or inclusions, and may therefore represent remnant outlines of a growth face.

Thirdly, there are meandering markings—tear lines—which display a fine-scale stepwise structure suggestive of micellar imperfection. Fourthly, a large whorl appears in the lower center of the field, which appears to nucleate at the site of an inclusion or void. This whorl pattern may refer to growth imperfection, although it may also result from stress resolution around the pit (Type II pattern). It is an interesting feature that some of the tear lines completely reverse their direction. Before assigning this whorl to stress, reference should be made to similar patterns in ferrosilicon which definitely express imperfection from compositional changes in the neighborhood of the inclusion or void (19, 20).

Lastly, immediately to the right of this inclusion a short straight marking shows itself heavily shaded. It lies parallel to the bands ascribed to twinning and therefore may represent *parting* on the twinning plane. A similar instance will be shown later in calcite. Careful inspection of the stepwise meandering markings, just listed as the third feature, will show that these steps have their most prominent direction parallel with the bands, which suggests that a finescale weakness has developed from minute twins not otherwise observable. Such fine twins or “striae,” of course, are well known for plagioclase.

In the lower fractograph of Fig. 6, a field is shown for plagioclase which has an especially pronounced fanlike pattern combined with good examples of twinning. It is tempting to relate these fanlike markings to growth imperfection, but a reservation must be held for the effect of stress pattern.

In concluding the discussion of plagioclase, attention will be called to a special study which the preceding fractographs invite—an investigation of possible pattern changes across the constitutional range of this isomorphous plagioclase series. A marked sensitivity has been demonstrated in this respect for fractographs of metallic systems (19, 21).

Orthoclase

Passing now to the potassium aluminum silicate, orthoclase, the fractographic pattern in Fig. 7 can be tentatively accepted as typical.

No twinning is in clear evidence, and this seems to constitute the principal fractographic difference between the specimens of orthoclase and plagioclase examined in this research. The wavy bands of tear-line families, however, seem to stem from approximately parallel markings of unknown origin. These latter may relate to twinning; but in work with cast molybdenum metal (14), and in studies with bismuth-antimony

alloys, markings of this type have been related to pulsating solidification fronts during growth of the crystal. They may therefore similarly relate here to the growth history of the crystal. The tear-line patterns are elaborate in this material, and may also reflect growth imperfection, although this matter will require much further study.

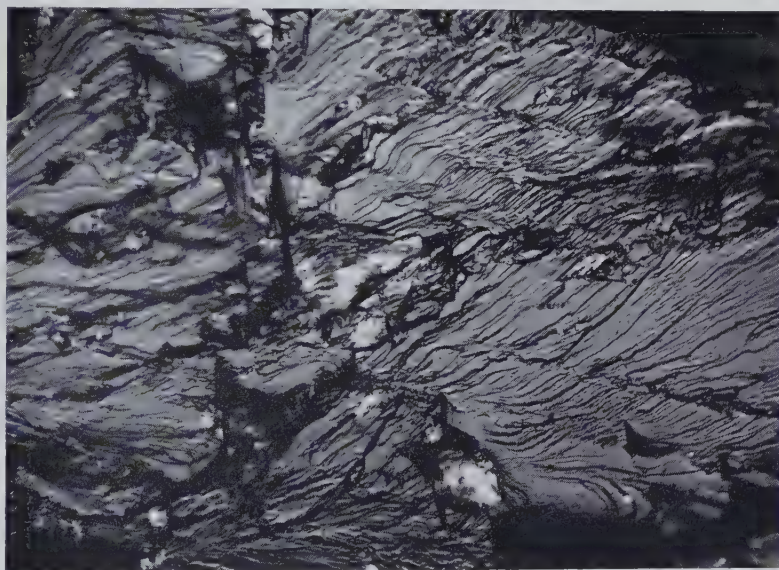


FIG. 7. Typical fractograph of orthoclase feldspar. 235X.

MUSCOVITE

Since the mica family provides a remarkable cleavage, a sample of muscovite was studied. The extremely perfect basal (001) cleavage renders the surfaces relatively free of intersecting markings; but Type Ic patterns may appear from interleaved phases.

In Fig. 8, two such patterns are shown. The upper fractograph discloses a number of very thin, transparent platelets trapped beneath the lamella of the cleavage facet. The focus is directed upon these, rather than upon the cleavage face itself. No analysis of these entrapped platelets was attempted; but their symmetry and transparency suggests that they are micaceous forms similar to the parent crystal.

In the lower fractograph of this figure, however, a beady phase appears which can readily be concluded to be foreign to the system. Again no analysis was attempted, the contribution resting with the fact that such phenomena are so clearly displayed.

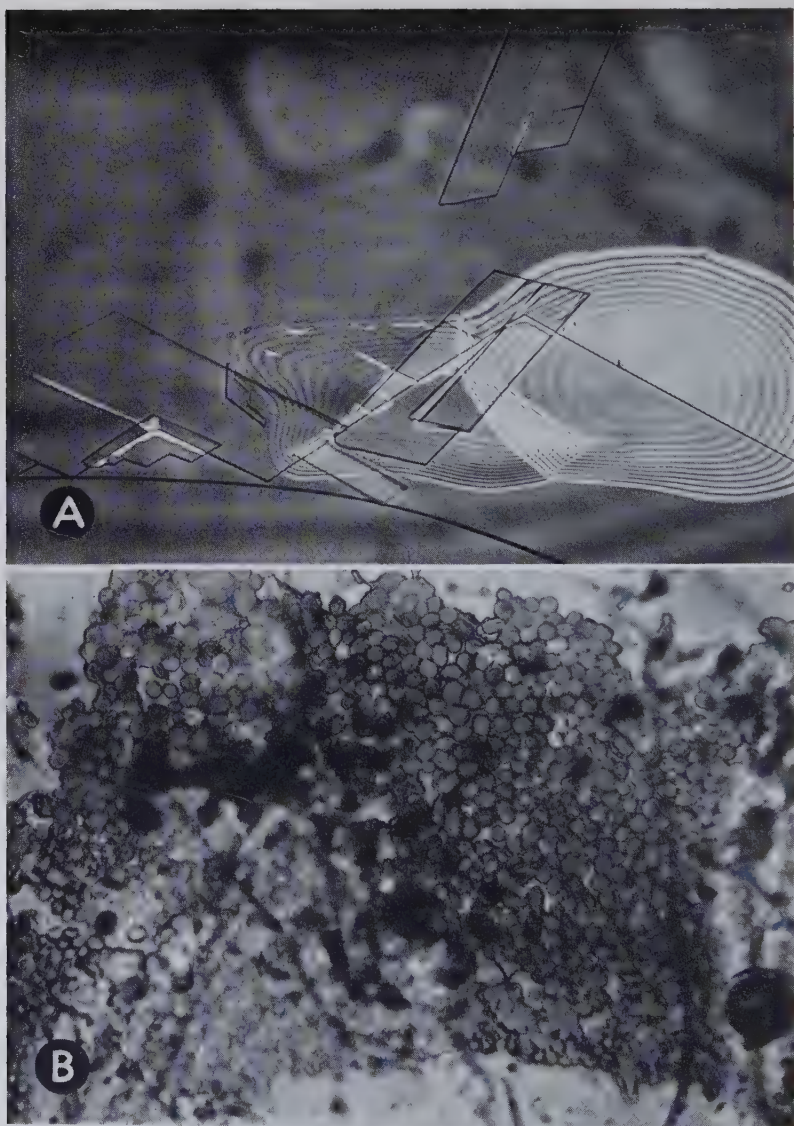


FIG. 8. Fractographs of muscovite. (Above) Interlamellar entrapment of minute crystals, probably of muscovite. 235 \times . (Below) Interlamellar entrapment of a foreign phase. 335 \times .

FLUORITE

Fluorite, CaF_2 , is an isometric mineral having such excellent cleavage on the octahedron $\{111\}$ and such transparency that the typical fractographic field is not particularly informative. Specimens from two different sources (Nos. 8 and 9 in Table 1) were studied; and a fractograph of each is presented in Fig. 9, principally for displaying some uncommon observations.

In the upper fractograph, one prominent intersecting cleavage appears, along with tear lines which are extremely regular in their disposition. This suggests that they conform to crystal weaknesses (Type I), rather than to stress resolutions (Type II). These weaknesses, of course, can be Type Ia crystallographic directionalism; but close observation of minute stepwise effects, both in a linear sense along tear lines and in a lamellar sense in the gradations of the cleavage levels, suggest a Type Ib mosaic effect.

In the lower fractograph, a most remarkable pattern appears. If this is attributed to growth imperfection of lineage or dendrite type, it is especially fascinating to find such an elaborate architecture within a matrix known to belong to a single crystal, hence having a common orientation; for it can only signify that the over-all molecular structure has maintained an approximately exact orientation, but that throughout the matrix there is distributed an imperfection pattern relating to crystal growth. The micellar theory accounts for this on the basis of a deposition of molecular clusters, or micelles, from the liquid state, such that the molecules within each micelle maintain the orientation common to the field, but a displacement is suffered among the micelles themselves. Fracture, strongly influenced by the path of maximum weakness, proceeds along the intermicellar boundaries and thereby depicts the imperfection architecture, of which this fractograph in Fig. 9 may be an excellent example.

THE CARBONATES

Once again a promising special study is suggested by the calcite group, whose various members clearly provide varying fractographic fractures. Here only a brief outline will be given.

Dolomite and Limestone

In Fig. 10, two fractographs are compared for dolomite and limestone, although the latter carries poor definition as a mineral. Nevertheless, both a relationship and a distinction are displayed. The dolomite shows the customary rhombohedral $\{10\bar{1}1\}$ perfect cleavage, with numerous

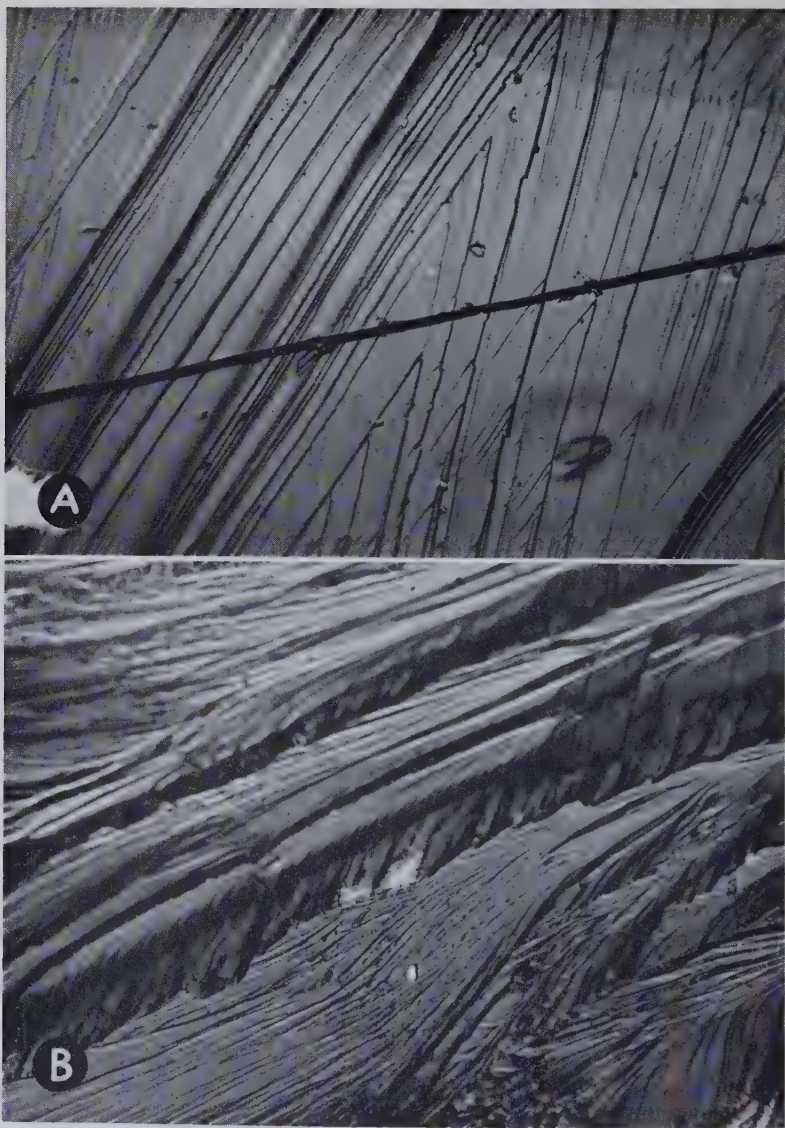


FIG. 9. Fractographs of fluorite. (Above) Intersecting cleavage and Type I tear lines. 235 \times . (Below) Phenomenal pattern believed to express growth imperfection. 315 \times .

bands or intersecting cleavages at angles closely approaching 90° . The limestone, on the other hand, shows only slight traces of these intersecting markings; and the cleavage is comparatively rough. This roughness shows itself as a mass of fan-like markings, or groups of tear lines; and

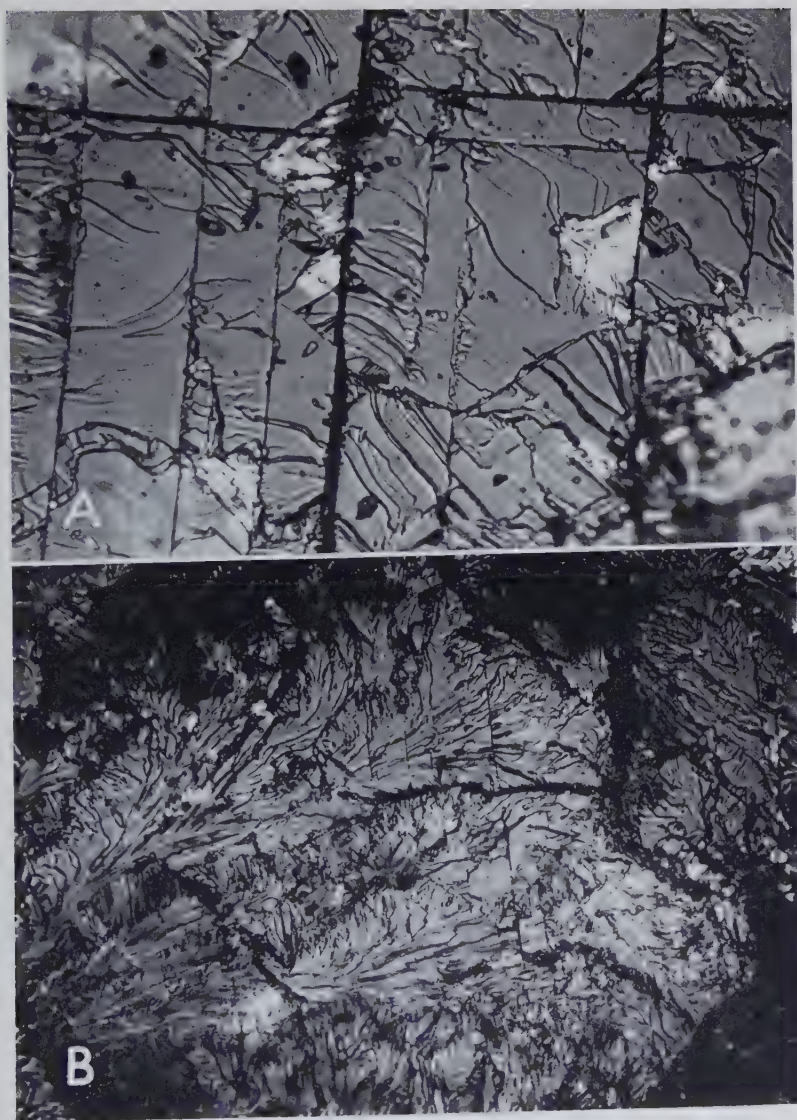


FIG. 10. Fractographs of dolomite (above) 335 \times , and limestone (below). 235 \times .

it is perhaps reasonable to conclude that these are structural patterns of growth imperfection, aggravated in this material by the greater complexity of its composition.

Calcite (Twinned)

Two samples of calcite were studied, one much twinned (No. 12 in

Table 1), and one optically pure (No. 13 in Table 1). Figure 11 presents a fractograph taken of the twinned sample No. 12 with the customary oblique illumination. Large tear lines or tear surfaces appear, overlain with an immense amount of fine detail. This detail somewhat divides itself into (1) a set of very delicate markings which are approximately parallel and vertical on the field, and which are strongly affected by the tear markings; and (2) a set of coarser markings which are also approximately parallel, running diagonally across the field and quite independent of the tear traverses. The angle between these two sets provides some similarity with the markings in the upper fractograph of fluorite in Fig.



FIG. 11. Fractograph of twinned calcite. 235 \times .

9. Still a third set can be found among the delicate traces, particularly in the upper right of the field; and these indicate a change of direction across the coarser markings consistent with the identification of the latter as twins.

In an attempt to decide whether the field contained a twin, a duplicate fractograph was taken using polarized light, though it will not be reproduced in the interest of conserving space. A light band then appeared running diagonally across the field from the base of the tear line on the right, but otherwise relating to nothing in the pattern. The diffuseness of this single illuminated band, however, suggested that it lay beneath the cleavage surface. Except for a uniform darkening over the whole portion of the distorted field, but little evidence appeared that any of

the pattern belongs to twinning. If twins are present, they must be fine superficial formations, formed at the instant of the formation of the new surface.

In Fig. 12, a fracture profile appears amid a pattern of tear lines and inclusions. Polarized light, in the lower fractograph, then reveals this profile to lie along a twin band; whereupon it can be presumed that the

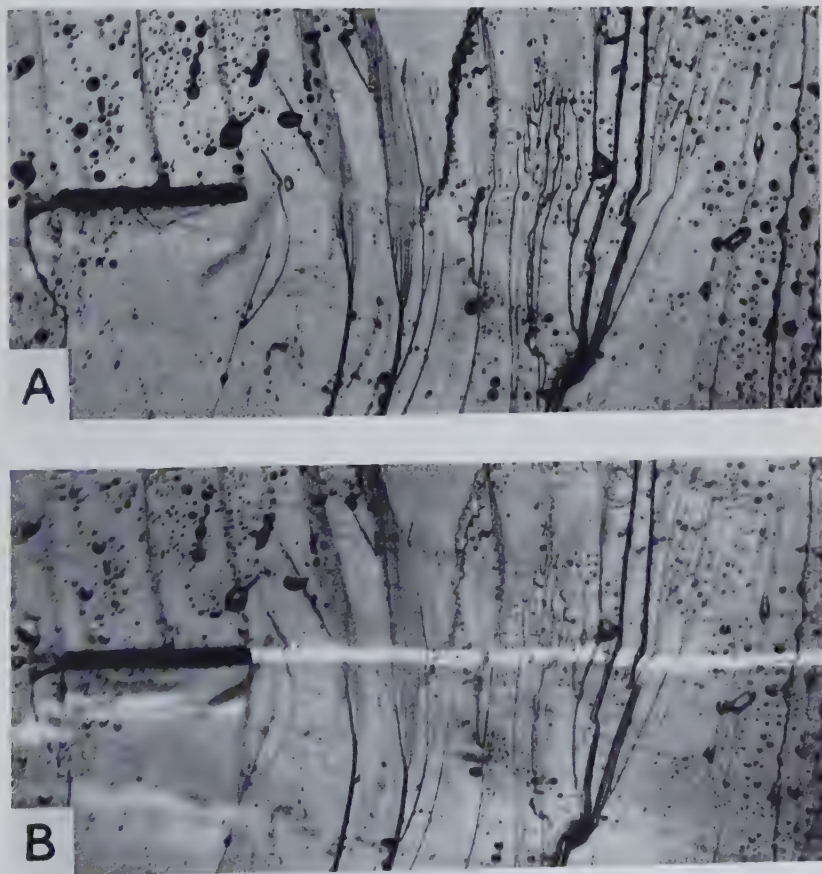


FIG. 12. Parting on a twinning plane in calcite. (Above) Oblique illumination. 350 \times .
(Below) Polarized light. 350 \times .

fracture represents parting on that twinning plane. Slight changes can be noted in the path of the tear lines as they cross this twin.

In Fig. 13, polarized light in the upper fractograph displays a prominent twin; and a rather remarkable displacement of this twin appears at the intersection with the heavy diagonal trace. Tear lines in the upper left of the field can be observed to deflect in crossing the twins; and very

close inspection of the fine striae in the lower right (perhaps difficult to observe in the reproduction) will show these similarly deflected. The lower fractograph in this figure displays a pattern which is quite remarkable, and not at all understood. Much of the detail in the central portion of

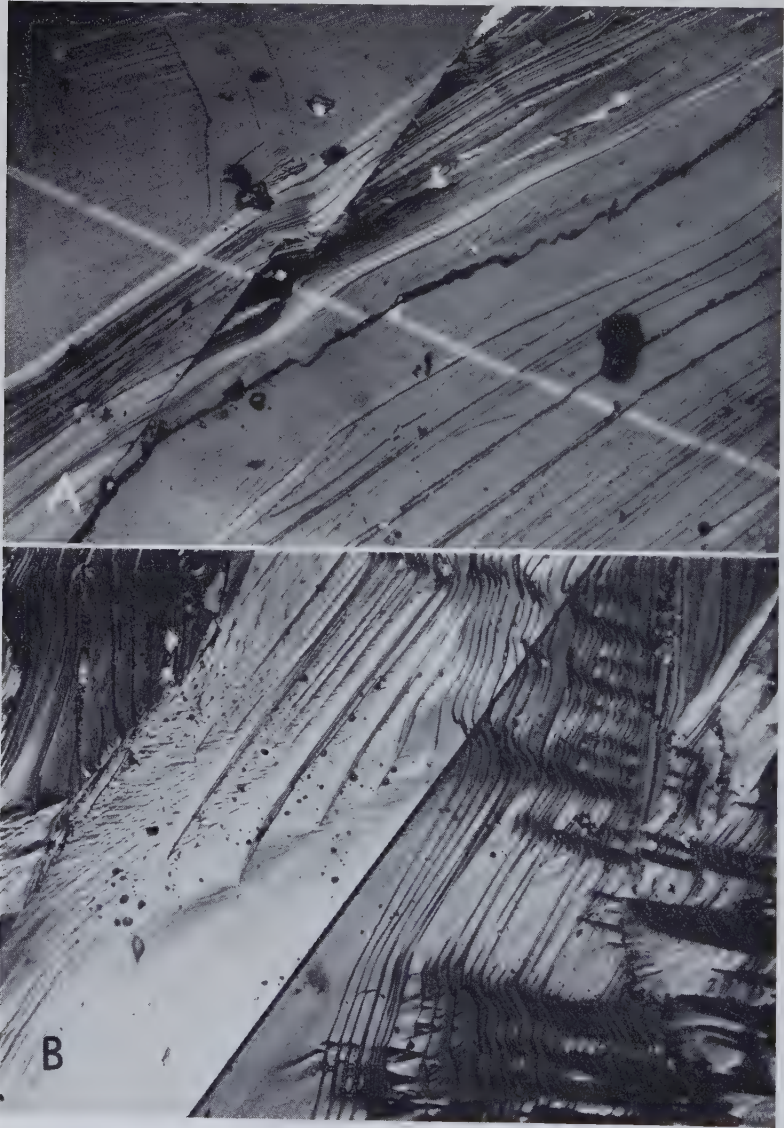


FIG. 13. Fractographs of twinned calcite. (Above) Polarized light. 235 \times . (Below) Oblique illumination of another field disclosing a phenomenal pattern. 235 \times .

the field, just to the left of the sharp diagonal line, is similar to the Type II markings illustrated in the previous Fig. 3; but the diagonal itself is puzzling, particularly because it simultaneously sections the field and yet imposes no alteration on the pattern at the upper right.

Four divisions can be noted in this fractograph, two to the left of the diagonal line, and two to the right. Counting these from left to right: (a) patterns 1 and 3 are similar, (b) pattern 4 shows strong evidences of twin structures, (c) none of the three boundaries dividing these fields



FIG. 14. Fractograph of optically pure calcite indicating lamellar imperfection. 235 \times .

is understood, and (d) fields 1, 3, and 4, particularly the latter, show strong indications of a lamellar imperfection structure, such as would derive from a micellar constitution.

Calcite (Optically pure)

By way of contrast, a laboratory sample of optically pure calcite (No. 13 in Table 1) was examined. The excellent cleavage provided visually flat fields, although at higher magnification considerable detail was exposed. The fractograph in Fig. 14 shows a field segmented by a diagonal boundary. The boundary shows a width, and the tear lines are short and markedly angular, disclosing the crystallographic directions to some extent. The surfacial lamellae outlined by these angular tear lines provide evidence for an inherent micellar imperfection structure even in this well developed crystal, and the matter should be explored

further from the standpoint of a possible relationship to optical and x-ray properties.

In Fig. 15, much evidence of fine-scale imperfection appears. The upper fractograph contains the same elaborate, delicate detail found in Fig. 11 for ordinary twinned calcite. This fine detail, then, can be due to twinning only if it develops during fracture. Strong indications of twinning stand in the chevron form of some of the markings; and the conclu-



FIG. 15. Fractographs of optically pure calcite revealing fine-scale lamellae and suggesting superficial twinning similar to the calcite in Fig. 11. (Above) 100 \times . (Below) 235 \times .

sion can perhaps be drawn that much of this pattern involves twinning. Nevertheless, it will be recalled from the previous discussion that crossed nicols gave no strong indication of reoriented material. As suggested earlier, perhaps only a thin layer is twinned, a micelle or so deep. This would account both for the evidence of twinning and for the inefficacy of analysis by polarized light, also for the fine lamellar detail of the pattern.

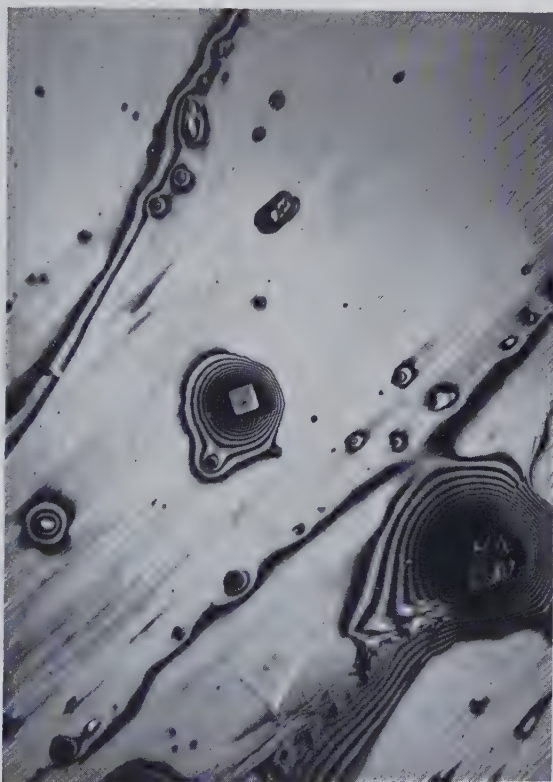


FIG. 16. Lamellar wall structure surrounding pits in optically pure calcite. 235 \times .

Attention is called to the particularly fine lamellar detail within the flat field at the top of the upper fractograph in Fig. 15. Several inclusions are also visible, two at left center apparently referring to a fluid phase.

In the lower fractograph of this same figure, a complex pattern is presented which reminds one of the previous Fig. 13 in the occurrence of several patterns within the one field. Most outstanding is the wavy formation in the center, which exhibits a lamellar form strongly suggesting micellar constitution. Note the columnar markings in this portion of the field approximately perpendicular to the heavy diagonal trace.

Markings in bismuth similar to these columnar ones are frequently observed, and their orientation similarly approximates a crystallographically significant position for that system (30° and 60°). Hence the present markings are not to be dismissed as Type II stress circumstances. It is also tempting to suggest that the tributary systems of tear lines in the lower left display the architecture of a cleavage cutting into the adjacent wave-like bank and following the indicated lamellar surfaces. This in turn raises the question why the field in the upper right, which is also visibly lamellar, exhibits a markedly different pattern. This cannot be answered by the present study; and the evidence will be left as material inviting further investigation. The thin straight trace across the top of the field should also be noted, for its surprising lack of deviation in crossing the several groups of markings shows the whole field of the fractograph to be quite flat.

As for the lamellar imperfection structure of calcite, even in the optically pure material, the concluding fractograph in Fig. 16 presents another aspect. Here pits appear, probably the seats of inclusions. The walls of these pits show a pronounced lamellar structure, hence one not influenced by deformation in this case, but by growth. The rings surrounding these inclusions are not to be confused with birefringence, such as occurs in the fractograph of muscovite in Fig. 8. The present rings are physical terraces which can be focussed upon.

CONCLUSION

From this preliminary study of several minerals and metals using the technique of fractography, the following conclusions can be tentatively drawn.

(1) Minerals, like metals, reveal a variety of detail upon their fracture facets which discloses structural features of the crystal.

(2) Fractographic patterns can be broadly classified as follows:

Type Ia: Patterns caused by crystallographic directionalism, whose traces therefore relate to the symmetry elements of the particular crystal;

Type Ib: Patterns caused by growth imperfection, or departure from the ideal directionalism of the crystal;

Type Ic: Patterns caused by extraneous or secondary phases;

Type II: Patterns caused by stress, resolving its waves and nodes in an effectively directionless matrix.

(3) All three subclasses of Type I disclose features of the crystal, and the patterns of both metals and minerals predominate in Type I.

(4) Type Ia discloses such crystallographic phenomena as (a) twin bands, (b) parting on twinning planes, (c) perfect or primary cleavage,

(d) intersecting primary and secondary cleavages, (e) unidentified striae perhaps representing fine-scale twinning, (f) markings possibly relating to historical growth faces, and (g) other markings.

(5) Type Ib refers specifically to crystal imperfection, which is an issue of much importance today. The fractographs of minerals, as of metals, provide abundant evidence for a mosaic or micellar constitution, particularly showing as fine-scale lamellae; and much evidence appears for lineage or dendrite-type growth imperfection during the aggregation of the micelles to form the crystal. This is true even of the more perfect crystals, such as optically pure calcite.

(6) Type Ic provides informative patterns for extraneous phases, specifically illustrated here for oxide and carbide in molybdenum, inclusions in numerous minerals, and two examples of entrapped inter-lamellar phases in muscovite.

(7) Type II only discloses features of the rupturing stress and is therefore more or less informationless so far as the crystal is concerned.

(8) Facets can be studied with any of the common types of reflected illumination; (a) vertical, (b) oblique, (c) dark-field, (d) sensitive tint, (e) polarized light, or (f) phase contrast. Oblique illumination is most useful, even for transparent minerals.

(9) Minerals can probably be readily distinguished from one another fractographically, as metals are, with further application of the technique; and some interesting special studies are suggested to determine, for example, whether members of an isomorphous series can be distinguished, since this has been accomplished for metals.

ACKNOWLEDGMENT

The authors acknowledge the sponsorship of the Office of Naval Research for a portion of this study; Dr. N. W. Buerger of the Naval Postgraduate School for consultation and contribution of specimens; and Dr. Grant M. Valentine, Geologist in the Department of Conservation and Development of the State of Washington, for his kindly interest and contribution of specimens.

APPENDIX

THE MICELLAR THEORY

One of the outstanding problems in researches on the solid state today is that of *imperfection structure*, and it is receiving first attention by laboratories in all parts of the world. Because fractographs directly disclose the internal structure of the crystal, a special significance attaches to the type of observations presented here. This Appendix is therefore added to clarify remarks in the text on *micellar structure*.

By "imperfection structure" is meant any phenomenon or disturbance which leads to a discontinuous atomic structure within an otherwise extended lattice. The fact of imperfection is no longer so much at stake, since most crystals are observably imperfect, but the inference from increasingly numerous observations is that crystals are by their very nature intrinsically imperfect. The history of the much involved argument on this subject can be found in Refs. 7, 8, 11, 12, and 16.

Briefly, it is now coming to be widely granted that crystals are minutely subdivided by discontinuities. Terminology for these discontinuities includes "dislocations," "Verhakungen," "Lockerstellen," "rifts," etc., whereas the structure itself is often referred to as "mosaic," signifying its subdivided nature.

In 1949, the senior author published a "micellar theory" which specifically accounts for the observed imperfection as the result of a cluster condition in the fluid state prior to solidification. If the theory is correct, a new field of research is opened for investigation of many important properties of solids not yet acceptably explained. Even if the theory is incorrect, the type of study promoted by its consideration is an important one.

TWELVE PRINCIPAL POINTS OF THE GENERAL THEORY

In brief topical outline of its principal aspects, the following twelve definitions summarize the general theory, from the particular standpoint of the solid inorganic state:

- I. That a universal condition of aggregation of atoms (or molecules) obtains within the liquid and/or gaseous states, probably at all temperatures, but certainly in a range just previous to solidification, this condition, because of its occurrence within an otherwise homogeneous constitution, being tentatively referred to as *isocolloidal*, and the aggregates as *micelles*.
- II. That the size of the individual micelle in the fluid state at any given temperature, other factors constant, approaches a preferred dimension expressing a balance among forces broadly represented as associative valence forces, dissociative thermal forces, and surface energy.
- III. That the shape of the individual micelle is a preferred crystal form for its system, having a high internal perfection modified externally by curvatures expressing the action of surface tensile forces, and perhaps further modified by adsorbed fractions of extrinsic phases in multicomponent systems.
- IV. That solidification has the nature of gelation, or agglomeration, of these micellar units—as opposed to the molecular concept

of solidification—accordingly causing the solid state to inherit a virtually permanent and discrete substructure discontinuous on a scale of colloidal dimensions.

- V. That the approximate matching with regard to crystallographic orientation of the micelles during solidification provides the gross structure known as the lineage, dendrite, grain, or crystal.
- VI. That the mismatching of the micelles during solidification provides the imperfection structure of the crystal which has attracted so much attention and so little agreement in discussions on the solid state.
- VII. That this imperfection structure of the solid state therefore constitutes a universal and as yet unavoidable feature of all crystals larger than the unit micelle, such that the best precautions now known for crystal growth can effect no more than an improved matching of the micellar individuals.
- VIII. That imperfections in the solid state are accordingly of three general classifications:
 - (a) *intermicellar*, which concern the fundamental fine-scale disjunctions between individual micellar faces.
 - (b) *interlineage*, which express the integrated intermicellar disjunctions occurring between two separately nucleated but commonly oriented aggregates or “lineages,” and
 - (c) *intergranular*, which relate to the surfaces of separately nucleated and separately oriented lineages or clusters of lineages (“grain” or “crystal”).
- IX. That the intramicellar forces are those primary molecular or valence forces which relate to the fundamental cohesive energy of the atoms in the perfect lattice structure, and upon which the so-called theoretical calculations of “ideal” crystal strength are based.
- X. That the intermicellar forces are principally secondary or adsorptive forces of markedly lesser magnitude, which determine the massive strength or cohesion of the “real” crystal and thereby explain the great discrepancy so characteristically noted between “real” and “ideal” crystals.
- XI. That the fundamental disjunction, the intermicellar adsorption face, upon which the entire imperfection structure of the solid state is thus based, involves a break in the regular lattice, not as vacant lattice sites predicated by most contemporary “dislocation” theories, but as a discrete and a highly persisting boundary for those micellar units from which it originates.
- XII. That these intermicellar boundaries provide (a) “slip planes”

for plastic deformation, whose minimum spacing is the unit micellar thickness, (b) cleavage surfaces, whose pattern is the fractograph, and (c) a universal presence of "internal surface" upon which all subsequent chemical and physical effects and constitutional changes must therefore be predicated.

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ALLANITE FROM YOSEMITE NATIONAL PARK, TUOLUMNE CO., CALIFORNIA

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ABSTRACT

Allanite-bearing rocks have been found in two localities in Yosemite National Park, Tuolumne County, California, *viz.* in narrow pegmatitic veins at Long Gulch, approximately one mile north-east from Aspen Valley, and in quartz-orthoclase pegmatite boulders in scree in the south slopes of Ragged Peak. Optical properties for these two occurrences are as follows: Long Gulch: $\alpha=1.769$; $\beta=1.784$; $\gamma=1.791$; $\gamma-\alpha=0.022$; $2V=60^\circ$ (—); dispersion is distinct with $r>v$; pleochroism is brown (Z) to pale yellowish brown (X); $D_{20}^\circ C=3.87$. Ragged Peak: $\alpha=1.791$; $\beta=1.815$; $\gamma=1.822$; $\gamma-\alpha=0.031$; $2V=40^\circ$ (—); dispersion is distinct with $r<v$; pleochroism is deep warm brown (Z) to pale brown (X); $D_{22}^\circ C=3.95$. Both minerals are readily decomposed by warm concentrated hydrochloric acid. Semiquantitative tests show that the Long Gulch allanite contains approximately 0.6 per cent MnO, whereas a complete quantitative analysis of the Ragged Peak mineral indicates that it contains 6.74 per cent and 2.02 per cent of MnO and TiO₂, respectively.

Both minerals exhibit only slight evidence of metamictization and both contain no lead. Acid treatment of allanite is discussed and it is shown that the degree of solubility is a function of composition and that true gelatinization will only result if the minerals are in a metamict condition.

PREVIOUS WORK

Allanite has been described from Yosemite National Park and adjacent country by Ries (1900), Pabst (1928, 1938), Calkins (1930), and others, but so far as the writer is aware neither analyses nor optical data are available for occurrences of this mineral from this general area.

LONG GULCH OCCURRENCE

The cerium-bearing epidote forms prismatic crystals that occasionally measure up to 15 mm. in length and these are sporadically distributed in narrow sheets of mineralogically simple pegmatite 3–4 inches in width. In general the allanite crystals are elongated parallel to the *b* crystallographic axis, and in sections that are cut parallel to a plane containing the *a* and *c* axes, a roughly hexagonal outline due to approximately equal development of basal planes, orthopinacoids, and hemiorthodomes is evident.

MINERALOGY OF THE PEGMATITE AND HOST ROCK

The host rock is a granodiorite with about 50 per cent of plagioclase that exhibits strong zoning of a normal oscillatory type, and a range of composition from An₃₂–An₃₉. However, in a number of crystals peripheral zones were observed that averaged in width about one-fifth of the grain diameters, in which there was little if any range of composition. Albite twinning is ubiquitous and is in most instances combined with twinning

according to the Carlsbad law. Where andesine adjoins orthoclase, narrow selvages show a myrmekite-like structure with minute vermicular interpenetration of plagioclase by orthoclase or microcline. Orthoclase, which makes up about 15 per cent of the total rock, is usually untwinned and here and there a few crystals of microcline with poorly developed cross-hatch twinning were observed. In the pegmatite itself microcline and orthoclase are pink in color and more plentiful than in the granodiorite; and in addition both potash feldspars exhibit a regular arrangement of inclusions of minute blebs of plagioclase (An_{26} ; $\alpha = 1.542$; $\gamma = 1.550$) of the same composition as the associated pegmatitic plagioclase, although the latter it will be noted, is less calcic than the plagioclase in the enclosing granodiorite. The potash feldspar of the pegmatite zone shows strong replacement structures where it adjoins the plagioclase of the host rock. In these instances large poorly twinned but irregularly shaped grains of microcline exhibit peripheral zones heavily studded with remnants or islands of calcic oligoclase (Fig. 1, *B*).

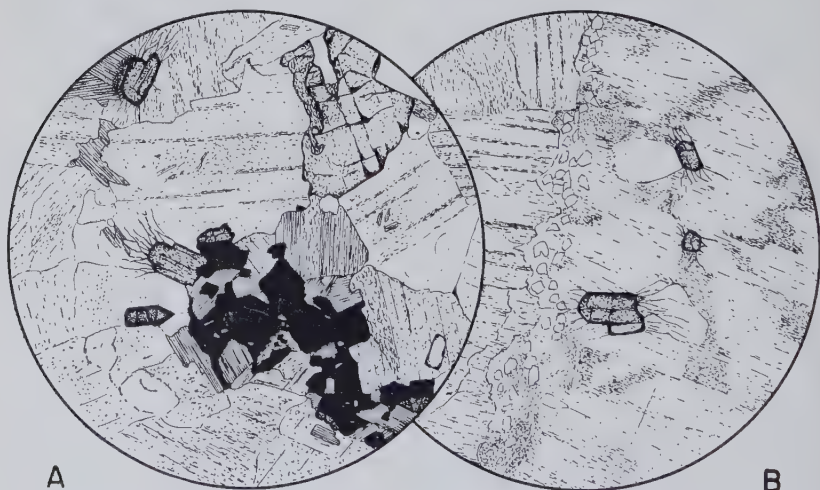


FIG. 1. *A*. Granodiorite showing allanite with cracks radiating therefrom, ferruginous epidote closely associated with a glomeroporphyritic segregation of titaniferous iron ore, chlorite, partially chloritized biotite, and sphenes, in addition to feldspars and quartz. A weak pleochroic halo has developed at the biotite-allanite junction in one case (top left-hand area). Locality: Long Gulch, Yosemite National Park, California. Magnification $\times 10$. *B*. Potash feldspar-oligoclase pegmatite with allanite; note the evidence of expansion in allanite crystals and the interpenetrative border associated with "islands" or relicts of plagioclase between microcline (right half of field) and oligoclase (left half). Locality: Long Gulch, Yosemite National Park, California. Magnification $\times 10$.

About 10 per cent of quartz is present in the host rock but it is more plentiful in the pegmatite, and whereas about 15 per cent of biotite is found in the former rock only occasional plates were found in the latter.

In the granodiorite, biotite is considerably altered to fine flaky chlorite, often in the form of tiny rosettes, and this transformation appears to have in some way facilitated the crystallization of ferriferous epidote and wedge-shaped crystals of sphene (average length 1.5 mm.), since these two minerals are frequently closely associated with the assemblage of chlorite and biotite (Fig. 1, A). Chlorite has the following optical properties:

$$\begin{aligned}\alpha &= \beta = 1.616 \pm 0.002 \\ \text{Birefringence} &= 0.002 \text{ (visual estimation)} \\ 2V &= 0^\circ (+) \\ X &= Y = \text{green} \\ Z &= \text{very pale yellow} \\ \text{Absorption: } X &= Y > Z\end{aligned}$$

These data suggest that the chlorite is an iron-poor member of the ripidolite group (Winchell, 1936, p. 649). Occasional crystals of allanite, iron-ores, apatite, and zircon complete the mineral assemblage.

Here and there the pegmatite band is intersected by narrow veinlets (10–20 mm.) that are composed entirely of radiating, finely prismatic, bright green crystals of iron-rich epidote.

ACCESSORY MINERALS OF THE PEGMATITE

The following constituents were recognized in the heavy residue separated from a two-pound sample of pegmatite by centrifuging the crushed rock in bromoform of density 2.88 *ca.*:—

Sphene. Sphene made up slightly more than one half of the total residue. It occurs as subhedral grains, and occasional “envelope”-shaped euhedra that exhibit prominent development of flat domes.

Iron oxides. Ilmenite and magnetite are both present in subhedral fragments, with the latter mineral exhibiting some peripheral oxidation to limonitic material.

Tourmaline. Only very occasional pale greenish-blue, fractured, prismatic crystals of tourmaline were observed.

Epidote. A few irregularly shaped fragments of yellow ferriferous epidote ($\gamma - \alpha = 0.025$ *ca.*) were found.

In addition the following minerals were observed as members of the heavy mineral assemblage although altogether they did not exceed approximately 5 per cent of the total residue: allanite; clear, colorless zircon; faintly pink garnet with $n = 1.825$; clear, colorless prisms of apatite devoid of inclusions; biotite; and monazite (one grain only). Finally one anhedral fragment of an unidentified golden-yellow isotropic mineral was observed which caused distinct darkening when placed on the emulsion of an α -ray track plate, and for which a refractive index of 1.650 was determined.

CHEMICAL PROPERTIES OF ALLANITE

The finely crushed mineral was completely decomposed, except for the production of pulverulent but not gelatinous silica, after treatment on the water-bath for five minutes with concentrated hydrochloric acid. Chemical tests showed that approximately 0.7 per cent of ThO_2 and 0.6 per cent of MnO is present, but no reaction for uranium was detected.

PHYSICAL PROPERTIES OF ALLANITE

Subhedral to euhedral crystals of allanite are distributed haphazardly throughout both pegmatite and granodiorite, but are more plentiful in the former. The allanite crystals are black, but crushed fragments exhibit a resinous to vitreous luster with pronounced conchoidal fracture. An imperfect cleavage parallel to the length of the crystal, presumably the basal cleavage, was observed in only a few instances.

The following optical data were obtained for Long Gulch allanite:

$$\alpha = 1.769 \pm 0.003$$

$$\beta = 1.784$$

$$\gamma = 1.791$$

$$\gamma - \alpha = 0.022$$

$$2V = 60^\circ (-) \pm 3^\circ$$

Dispersion: Distinct with $r > v$.

Pleochroism:

X = pale yellowish-brown

Y = dark brown

Z = brown

Absorption: $Y > Z > X$

$$D_{20^\circ\text{C.}} = 3.87 \pm 0.02 \text{ (torsion balance determination).}$$

Although the optical properties of Long Gulch allanite appear to suggest an unaltered mineral, incipient change towards a metamict condition appears to have commenced because a study of thin sections reveals a delicate system of anastomosing cracks that radiate away into the surrounding feldspar from several allanite crystals (Fig. 1, *A* and *B*). The cause of the development of the radiating fissures in such instances has been discussed by several writers (Ellsworth, 1922, p. 58; 1927, p. 367; Walker and Parsons, 1923, pp. 27-28; Hutton, 1950*A*, pp. 695-697).

RAGGED PEAK MANGANOAN ALLANITE

OCCURRENCE

At this locality roughly shaped crystals of manganoan allanite that measured up to 45 mm. in length and exhibited only traces of any development of crystal faces, were found embedded in blocks of quartz-orthoclase pegmatite. Rocks of this type made up a considerable propor-

tion of a coarse scree that had accumulated on the lower slopes of Ragged Peak and a search for occurrences *in situ* were unsuccessful, but nevertheless these must be near.

CHEMICAL PROPERTIES

Large anhedral fragments of allanite were readily separated from matrix in the first instance by hand-picking of coarsely crushed pegmatite. This concentrate was then crushed to pass a 230-mesh screen, freed of dust by careful elutriation and a sample of relatively homogeneous material was finally obtained by electromagnetic fractionation of the elutriated material by means of a Frantz Separator. With the Separator adjusted at a slope of 15° , a tilt of 5° , and a current of 0.10 ampere the attracted portion consisted chiefly of opaque iron oxides, and uniaxial, possibly oxidized, allanite. At 0.15 ampere and the same setting additional oxidized allanite and allanite with iron oxide inclusions were obtained, whereas at 0.20 ampere a nearly pure homogeneous allanite sample separated. This material was ground finely and centrifuged in Clerici solution over a range of densities to ensure purification of allanite from traces of cryptocrystalline quartz and iron oxides. The altered material and foreign particles did not exceed approximately 3 per cent of the unfractionated and uncentrifuged mineral fragments.

The analysis of this material (Table 1, analysis A) is interesting in several respects, *viz.* the high content of iron, mainly in the higher valency state, titanium, and particularly manganese as Mn^{2+} . The low percentages of SiO_2 and combined H_2O , and an absence of K_2O and Na_2O are indicative of the purity of the sample as analyzed. Analyses that are comparable in every respect to that of Ragged Peak manganian allanite have not been found, since allanites with a high content of manganese, either as Mn^{2+} or Mn^{3+} appear to be uncommon, and furthermore, allanites combining a high percentage of ferric iron, manganese, and titanium appear to be decidedly rare. However, so far as these three constituents are concerned a mineral from the Vishnevy Mountains described by Ovchinnikov and Tzimbalenko (1948) as mangan-orthite (Table 1, analysis D) is rather similar to the Ragged Peak occurrence, although the latter differs on account of the higher manganese content, the lower alumina, and the presence of both uranium and thorium. Unfortunately the constituents in the rare earth group in analysis D have not been differentiated. From the point of view of the content of Al^{3+} , Fe^{3+} , and Ti^{4+} in particular, the Ragged Peak mineral is remarkably similar to allanite from Colorado (Table 1, analysis F) described by Goddard and Glass (1940, p. 398) and furthermore, the optical properties of these two occurrences are almost identical.

Accordingly Ragged Peak allanite appears to contain a higher percentage of both manganese and titanium than in other analyses so far reported and this unusual composition taken in conjunction with the high figure for Fe^{2+} and Fe^{3+} , and particularly the latter, undoubtedly accounts for the unusually high values recorded for the refractive indices and birefringence of this mineral.

When the analysis of Yosemite manganoan allanite is recalculated (Table 2) on the basis of 13 (O, OH, F) atoms to the unit cell it conforms

TABLE 1. ANALYSES OF ALLANITES

	A.	B.	C.	D.	E.	F.
SiO_2	28.80	30.60	32.93	29.60	30.76	30.40
Al_2O_3	10.00	16.18	14.51	14.34	13.50	10.25
Fe_2O_3	10.25	3.52	nt.dt.	8.38	3.87	10.33
FeO	7.33	8.60	15.23	9.38	12.21	10.29
TiO_2	2.02	0.36	nt.dt.	1.96	0.13	1.46
MgO	0.58	0.46	0.74	0.82	0.45	1.44
MnO	6.74	4.48	4.32	5.37	4.87	0.66
CaO	10.08	7.76	12.61	10.56	7.35	7.47
Na_2O	nil	nil	nt.dt.	nt.dt.	nt.dt.	0.02
K_2O	nil	nil	nt.dt.	nt.dt.	nt.dt.	0.16
ThO_2	0.95	1.02	1.64	nt.dt.	2.26	nil
UO_2	0.015	nt.dt.	trace	nt.dt.	nil	nt.dt.
Ce_2O_3	9.06	19.36	4.87	17.20	22.82	14.61
La_2O_3	13.36	nil	8.19	nt.dt.	nt.dt.	10.32*
Y_2O_3	nil	2.22	nt.dt.	nt.dt.	nt.dt.	nt.dt.
$\text{H}_2\text{O}+$	1.32	5.07	4.58	1.08	1.36	1.95
$\text{H}_2\text{O}-$	nil	0.22	nt.dt.	nt.dt.	0.31	nt.dt.
	100.505	99.85	99.73	99.68	100.24	99.36

* The figures 10.32 are simply listed as "other rare earths" by the analyst.

- A. Manganoan allanite from Ragged Peak, Yosemite National Park, Tuolumne County, California. Analyst: Johnson, Matthey. The following constituents were looked for but found to be absent: ZrO_2 , V_2O_5 , P_2O_5 , Zn, Ni, Co, Pb, Cu, Ag, Cr_2O_3 .
- B. Allanite from Woodstock, N. W. Division, Western Australia. Analyst: D. G. Murray (Simpson, 1931).
- C. Allanite from Ishikawa, Iwaki Province, Japan. Total includes: VO_2 tr., SnO_2 tr., CO_2 0.11 (Kimura, 1925).
- D. Mangan-orthite from Vishnevsky Mts., Russia. Total includes: ZnO 0.72; V_2O_5 0.27. Analyst: I. B. Petropavlovsky (Ovchinnikov, L. N., and Tzimbaleiko, M. N., 1948, p. 192).
- E. Allanite from Ryôzen, Japan. Total includes: SnO_2 0.13; P_2O_5 0.07; CO_2 0.15; Y_2O_3 included with Ce_2O_3 . Analyst: S. Hata (Hata, 1939, p. 116, table 2, analysis 2).
- F. Allanite from southern cerite deposit, near Jamestown, Colorado. Analyst: C. Milton (Goddard and Glass, 1940, p. 398, table 4).

TABLE 2. RECALCULATION OF ANALYSIS OF MANGANOAN ALLANITE

	Wt. per cent	(O, OH)	Metals	
SiO ₂	28.80	.960	2.874	} 3.00
Al ₂ O ₃	10.00	.297	1.185	
TiO ₂	2.02	.050	.149	} .126
Fe ₂ O ₃	10.25	.192	.766	
FeO	7.33	.101	.605	} 1.059
MgO	0.58	.014	.084	
MnO	6.74	.094	.563	} .337
CaO	10.08	.180	1.077	
ThO ₂	0.95	.006	.018	} .226
Ce ₂ O ₃	9.06	.081	.323	
La ₂ O ₃	13.36	.123	.491	} 2.135
H ₂ O	1.32	.073	.874	

The formula derived is: (Ca, Th, Ce, La, Mn)_{2.13}(Al, Fe''', Fe'', Mg, Mn)_{3.00} [(Si, Al)₃O₁₂](OH)_{.87}

closely to the formula $X_2Y_3Z_3(O, OH, F)_{13}$ developed by Machatschki (1930, p. 96) for allanite, in which the X-group includes cations with ionic radius about $1.0 \text{ \AA} \pm 0.10$, the Y-group ions of intermediate size, and z, silicon, although aluminum and rarely beryllium may be present in the tetrahedra. In this connection three points deserve comment: (1) The low figure for Si⁴⁺ is balanced by an appreciable quantity of aluminum in the silicon-oxygen tetrahedral groups;* it should be observed that four-fold coordination of aluminum, normal in a great many silicates, is usually of little moment in allanites as Lokka (1935, pp.11-12) has made clear for some Finnish occurrences. (2) In order to satisfy the requirements of the structural formula manganese has been divided between the six-fold coordinated group and the X-group of higher coordination, a mode of distribution that is permissible owing to the intermediate size of the manganese ion. Machatschki (1930, p. 91) has included manganese in the X-group of his generalized formula, although in one of the analyses recalculated by him (Machatschki, 1930, p. 93) distribution of manganese between the X- and Y-groups was found to be necessary; this is in conformity with the circumstances usually found in

* It is interesting to note that Petropavlovsky and Tzimbalenko (1948, p. 192) have chosen to group Ti⁴⁺ with Si⁴⁺ in the tetrahedral assemblage but not Al³⁺.

piedmontite (Machatschki, 1930, p. 89). (3) The figure for hydroxyl is slightly less than theoretical requirements and on this account it was suspected that fluorine might be present; however, tests for the halogen were negative.

The reaction of the finely powdered mineral with acid is comparable to the behavior of the Long Gulch mineral under similar circumstances, *viz.* complete decomposition and solution except for the separation of insoluble, non-gelatinous silica, after boiling in concentrated HCl for about five minutes.

PHYSICAL PROPERTIES

The roughly shaped and dominantly anhedral crystals are black in color and when broken exhibit subconchoidal fractures that have a decided pitch-like aspect. A distinct cleavage is evident on broken surfaces in hand specimen.

In thin sections cut from large crystals of manganoan allanite zoning and heterogeneity were observed although the former development was in the main restricted to narrow and intermittent border phases in its distribution. Cleavage, believed to be parallel to {001}, is often very well developed in the peripheral zones, but it is indistinct and interrupted in the main part of any crystal, that is in the unaltered allanite. The peripheral zones are generally pale brown in contrast to the deep red-brown color of the phase which makes up the main body of the crystals, and they differ in other respects as well (Table 3). In turn the pale brown modification of the normal allanite is outwardly but patchily fringed by colorless clinozoisite (Fig. 2A). In the border areas chalcedonic quartz and opaque iron oxides occur (Fig. 2A) and here and there aggregates of diminutive and almost euhedral crystals of albite ($\alpha' = 1.526$) that exhibit twinning according to the albite law. Blebs and lenses of a golden yellow to red-brown phase occur throughout much of the border areas of the crystals (Fig. 2B). In some instances there is no clearly defined preferred orientation of this modification, whereas in others it forms narrow ribbon-like areas arranged parallel to the {001} cleavage as well as less regularly shaped patches that appear to follow fissures; and finally it occurs in rectangular and dart-like shapes, arranged *en echelon*, or very occasionally distributed along a curious linear feature with re-entrant angle (Fig. 2B). The optical properties of the golden yellow to red-brown phase are listed in Table 3, but unfortunately the writer is unable to show what the exact nature of this modification is. With some diffidence it is suggested that it may represent the combined effects of incipient metamictization and oxidation of allanite and its distribution in most instances along channels that would allow relatively easy access for solutions gives

some support to this suggestion. The refractive indices are distinctly lower but the birefringence is higher than the figures obtained for unaltered allanite. Undoubtedly transformation to a metamict condition would ordinarily tend to produce a product with lower refractive indices and birefringence, but on the other hand oxidation of both manganese and iron would be likely to increase the value of these figures, and there are large amounts of these metals in lower valency states in the unaltered



FIG. 2. *A.* Narrow altered and zoned peripheral area in large crystal of manganoan allanite. Areas exhibiting cleavage consist of pale colored allanite with large optic axial angle, and clinozoisite. Note the ragged grains of magnetite set in a patch of chaledonic quartz. The relatively homogeneous areas are brown allanite with poorly developed cleavage and occasional patches and blebs of red-brown uniaxial allanite (the less dense areas). Clear patches are holes in, and edge of, thin section. Locality: Ragged Peak, Yosemite National Park, California. Magnification $\times 25$. *B.* Thin section of manganoan allanite showing junction with quartz of pegmatite vein and distribution of blebs of red-brown uniaxial allanite. Note also the unusual linear feature, as yet unexplained, and distribution of wedge-shaped areas of uniaxial phase along that direction. Locality: Ragged Peak, Yosemite National Park, California. Magnification $\times 5\frac{1}{2}$.

allanite (*vide* Table 1, anal. *A*). The cumulative effects on the optical properties of allanite of these two processes acting simultaneously but to different degrees of completeness cannot be adequately assessed in this case.

The linear feature already mentioned (Fig. 2*B*) does not seem to be the result of twinning because a careful check with the universal stage showed no change whatever in the positions of any element of the indicatrix in the mineral on either side of this boundary. Nor does it seem likely that it is the result of an abrupt change in chemical composition,

because the optic axial angle is constant and in allanite 2V is fairly sensitive to compositional changes. Accordingly the writer is unable to offer a satisfactory explanation for the linear feature at this time.*

In accordance with Marble's (1937, p. 66) recommendation a radiograph of manganoan allanite was made, using a large crystal on which a carefully polished surface measuring 35×10 mm. was prepared. This surface was cleaned and then placed on a Kodak nuclear track plate type NTB for twenty-one days after which the plate was developed in Kodak solution D-8. The radiograph very clearly demonstrated the lack of homogeneity in the crystal as a whole because slightly greater density was apparent in those areas of the plate adjacent to the golden yellow to deep red-brown modification than in the emulsion that was in contact with the homogeneous deep brown allanite, that is, the analyzed material. In addition three very dense spots were found on the plate that indicated points of intense radioactivity, but a study of the thin section subsequently cut parallel to the polished surface did not reveal any obvious source for the radioactive particles that produced the over-exposed spots.

The following physical properties were determined for Ragged Peak allanite and its variants:

TABLE 3. OPTICAL PROPERTIES OF MANGANOAN ALLANITE AND ITS PHASES

	Pure analyzed allanite	Peripheral zone	Blebs
$\alpha =$	1.791 ± 0.005	1.779*	1.729*
$\beta =$	1.815	—	—
$\gamma =$	1.822	1.797*	1.770*
$\gamma - \alpha =$	0.031	0.018	0.041
2V	$40^\circ \pm 2^\circ$	$80^\circ \pm 2^\circ$	0°
Sign.	negative	negative	negative
Dispersion	$r < v$, distinct	$r < v$, distinct	—
Pleochroism:			
X =	pale brown	pale yellowish- brown	golden yellow to brown
Y =	brown	pale brown	—
Z =	deep warm-brown	pale brown	deep red to red-brown
Absorption =	$Z > Y > X$	$Z \geq Y > X$	$Z > X$
$X \wedge c =$	20°	27°	—
$D_{22^\circ C.} =$	3.95 ± 0.02	—	—

* A slight range of values was observed for the refractive indices and the data cited are averages.

* It should be noted that Russell (1937, pp. 1328-1329) has described an occurrence of twinning in allanite but gave no crystallographic data beyond stating that extinction was inclined to the composition plane.

The refractive indices recorded here appear to be distinctly higher than any previously recorded for allanite and undoubtedly they are a reflection of the unusually high percentage of Fe^{3+} , Mn^{2+} , and Ti^{4+} in the mineral. The only instance of allanite with comparable optical properties appears to be allanite described by Goddard and Glass (1940, p. 398) from the southern cerite deposits near Jamestown, Colorado, which as pointed out earlier, contains an unusually high content of Fe^{3+} and Ti^{4+} .

At this juncture it would be pertinent to call attention to the attempt made by Hata (1939, pp. 124-128) to correlate chemical composition with optical properties and particularly to an analysis of allanite from Kido, Japan (Hata, 1939, p. 127, Table 8, analysis 13). Hata plotted the percentages of $\text{Fe}^{2+} + 2\text{Fe}^{3+}$ in the *Y*-group against the percentage of rare earth elements in the *X*-group and some relationship would seem to exist between these data and the beta refractive index of the minerals concerned. Now the value of this relationship is, according to Hata, confirmed because the plot of the appropriate chemical data of an allanite with an unusually high percentage of Fe^{3+} (Hata's analysis 13) falls in his diagram on the correct refractive index line. However, the present author is of the opinion that this is entirely coincidental. If this particular analysis is treated as was done by Machatschki (1930) it is found not to agree with the formula $\text{X}_2\text{Y}_{3z_3}(\text{O}, \text{OH}, \text{F})_{13}$ but instead the *X*- and *Y*-groups are 1.52 and 3.46, respectively, if manganese is placed with the ions of higher coordination, or 1.39 and 3.58 respectively, if manganese is placed in six-fold coordination. Clearly then the allanite used for analysis by Hata was either impure or considerably oxidized, or alternatively the material employed for determination of the beta refractive index was not representative of the composition as stated in the analysis. When the appropriate data for Ragged Peak allanite are plotted on Hata's diagram it would appear that this mineral should have a beta refractive index of 1.775, a value that is very clearly distinct from that determined on material used for the purposes of analysis, *viz.* 1.815. And if Goddard and Glass's allanite is treated in the same fashion the value for beta as determined from Hata's diagram should be 1.799 instead of their value of 1.810. Clearly then factors other than those considered by Hata must be taken into account but these will be discussed in a later paper.

AGE OF YOSEMITE PARK ALLANITE

Although both thorium and uranium have been found in manganoan allanite and thorium only in the specimen from Long Gulch, lead was not detected in either specimen in spite of the fact that the host rocks, the granodioritic intrusives of the Sierra Nevada Range, are Jurassic in age. In a recent paper the writer (Hutton, 1951) has discussed a similar condition in allanite from New Zealand. In this case determina-

tion of the age of the allanite by the "lead-ratio" method gave an approximate figure only but certainly one that could not have exceeded $14-15 \times 10^6$ years although the mineral was unaltered and found in rocks that are of late Paleozoic age. In like manner the Californian allanites described in the present paper exhibit little transition towards a metamict state, a condition that has been tested chemically as well as optically; therefore, it appears reasonable to assume that there has been little or no loss of radiogenic lead by solution.

In an attempt to explain the wide discrepancy between the age determined by the "lead-ratio" method and the age of the host rocks determined by geologic field studies, the writer advanced the view that the New Zealand allanite is the product of very late redistribution and recrystallization of original pegmatitic allanite by solutions acting along old pegmatite or other solution channels; in other words, it is suggested that allanite may be as mobile a mineral as are members of the clinozoisite-epidote group. In the absence of any indications to the contrary it is suggested that similar conditions, *viz.* solution, redistribution, and resegregation at a fairly late date, could explain the absence of lead in these Californian occurrences. This view is not intended to infer that allanites are generally valueless for purposes of age determination since many examples are available in which the "lead-ratio" agrees satisfactorily with geological field evidence (Marble, 1935, 1943), although in other instances (Marble, 1940) it has been quite unsatisfactory. Finally, it must be borne in mind that allanite need not necessarily contain any thorium or uranium (Marble, 1942, p. 63), and such specimens are naturally useless for age determination by the "lead-ratio" method.

EFFECT OF CONCENTRATED ACID ON ALLANITE

When fine powders of Long Gulch and Ragged Peak allanites are heated with concentrated hydrochloric acid on the water-bath, solution is complete within about five minutes except for the separation of pulverulent but not gelatinous silica. Nor did gelatinization occur when powdered allanite was first ignited to dull-red heat for five minutes before acid treatment. This is not in accord with the behavior suggested for allanite (Murata, 1943, pp. 552, 555), since it is an orthosilicate, but nevertheless it does seem to be similar to that recorded for nagatelite, a phosphorian allanite. However, the reaction of acid on the Yosemite allanites is in marked contrast to the behavior of allanite from Wilmot Pass, Fiordland, New Zealand, under similar circumstances (Hutton, 1951). These distinctions in behavior invite some investigation and discussion.

First of all so far as the behavior of allanite in concentrated acid is concerned it would seem to be necessary to differentiate between meta-

mict and nonmetamict varieties. To check this it was found that the fine powders of two metamict allanites* from different localities underwent rapid decomposition when treated with hot concentrated hydrochloric acid, and provided the volume of acid initially employed was small the solution rapidly set to a thick rigid jelly. However, if these metamict allanites are heated first to bright-red heat for about five minutes and then treated with acid, decomposition of the minerals still occurred but in this instance the silica which separated was pulverulent and did not set to a thick jelly. Now the two allanites from Yosemite National Park exhibit only very slight indications of metamictization whereas the mineral from Wilmot Pass, Fiordland, New Zealand, shows no evidence of this transformation; the high birefringence and normal density determined for these minerals would seem to support this observation. Yet the Yosemite allanites are readily soluble in concentrated acid with separation of pulverulent silica whereas the New Zealand mineral is only slowly decomposed after the most drastic treatment, *viz.* prolonged heating with fuming sulfuric acid or perchloric acid. Unfortunately the composition of the allanite from Long Gulch is not exactly known except that the high refractive indices suggest a fairly ferriferous condition. Fortunately we can compare the composition of manganoan allanite from Ragged Peak with that of Wilmot Pass allanite (Table 4) and the outstanding and most significant differences between these analyses are the high percentage of octahedral positions occupied by iron and manganese in Ragged Peak allanite on the one hand and the dominance of aluminum in the octahedrally coordinated positions in Wilmot Pass allanite on the other. Now it is well founded that substitution of octahedrally coordinated aluminum by iron and (or) manganese leads to structural weakening which results in greater ease of chemical solution. This is found, for example, in andradite as compared with grossularite or spessartite; and lepidomelane as compared with muscovite among the sheet structures or disilicates.

However, it must be borne in mind in this connection that so far as orthosilicates are concerned other factors than replacement of six-fold coordinated aluminum by iron are undoubtedly of importance. For instance, clinozoisite and iron-rich epidote are both equally insoluble in

* The allanites employed in these tests were found at Albany, Wyoming, and Barringer Hill, Llano Co., Texas; the Wyoming mineral, which was not completely isotropic, although the birefringence did not exceed 0.005, has an average refractive index of 1.718, whereas for the Texas mineral a range of 1.718–1.724 was found. The composition of the Texas allanite must exhibit a distinct range since the refractive index recorded for a specimen from that locality by Glass (*vide* Marble, 1940, p. 172) lies outside the range of the values determined by the writer.

acid, and accordingly accurate determination of ferrous iron is notoriously difficult. However after preliminary ignition these epidote group minerals are readily decomposed by acid supposedly with the separation of a gel according to most texts. To verify this condition two clinozoisites ($\beta=1.709, 1.712$) and two ferriferous epidotes ($\beta=1.750, 1.755$) were ignited at bright red heat for five minutes and then treated with boiling concentrated HCl for a similar period. Although decomposition was rapid

TABLE 4. ANALYSES OF ALLANITES

	A.		B.	
Si ⁴⁺	2.986	} 3.00	2.874	} 3.00
Al ³⁺	1.871		1.185	
Fe ³⁺	.267	} 3.06	.766	} 3.00
Fe ²⁺	.796		.605	
Ti ⁴⁺	.050		.149	
Mg ²⁺	.095		.084	
Mn ²⁺	.016	}	.563	} 0.337
Ca ²⁺	1.192		1.077	
Th ⁴⁺	.022	} 1.89	0.018	} 2.13
Ce ³⁺	.301		.323	
La ³⁺	.323		.491	
Y ³⁺	.034		—	
H ⁺	1.169	1.169	.874	0.874

A. Allanite from Wilmot Pass, Fiordland, New Zealand (Hutton, 1951).

B. Manganoan allanite, Ragged Peak, Yosemite National Park, Tuolumne County, California.

a gel, that is a thick rigid jelly, did not develop even after evaporation of liquid almost to the point of dryness but instead a globular form of silica separated that readily settled to the bottom of the test tube. Since both clinozoisite and ferriferous epidote reacted towards acid in a similar manner, replacement of octahedrally coordinated aluminum by iron clearly produced little change in the stability of the ferriferous variety, whereas loss of hydroxyl through ignition must have caused a breakdown of the structure in both minerals with the result that solution in acid was easily accomplished.

The nature of the reaction of allanites with hot concentrated acid may be summed up as follows:

- (1) Metamict varieties: rapid production of a stiff jelly of gelatinous silica.
- (2) Non-metamict varieties:
 - (a) When the octahedral positions in the lattice are dominantly occupied by aluminum the mineral appears to be insoluble or only slowly decomposed after prolonged and drastic acid treatment.
 - (b) When the octahedral positions show considerable replacement by iron and (or) manganese the mineral is readily decomposed with the separation of pulverulent silica.

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STUDIES OF URANIUM MINERALS (VII): ZEUNERITE*

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ABSTRACT

X-ray study by the Weissenberg method on zeunerite from Tintic, Utah, gave a space group $P_4/nmm-D_{4h}^7$ and cell dimensions $a_0=7.13 \text{ \AA}$, $c_0=8.83 \text{ \AA}$, with $a_0:c_0=1:1.238$. The unit cell contents are $\text{Cu}(\text{UO}_2)_2(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}$. The calculated specific gravity is 3.79; the measured specific gravity of Schneeberg material is 3.64.

Study of synthetic zeunerite indicated the existence of two and possibly three hydrates, one with 16 to $10\text{H}_2\text{O}$, one with 8 to $5\text{H}_2\text{O}$, and possibly a still lower hydrate. In this respect the synthetic material is similar to autunite and torbernite, both of which have been found in the fully hydrated form and as lower hydrates, meta I and meta II. Natural zeunerite corresponds in water content and x-ray structure to meta-autunite I and to synthetic zeunerite with $8\text{H}_2\text{O}$.

Variation in water content affects both the optical and the fluorescent properties of the material.

Zeunerite, from the Weisser Hirsch mine near Schneeberg, Saxony, was named and first described by Weisbach (1872, 1873, 1877). Since then the mineral has been reported (Hintze, 1931) from at least seven other localities. The only available analysis of the Schneeberg material, made by Winkler (1873), established the composition as $\text{Cu}(\text{UO}_2)_2(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}$.

The mineral occurs as pale-green to emerald-green tetragonal crystals which usually are rectangular tablets flattened on $\{001\}$. The crystal habit closely resembles that of the isostructural mineral torbernite, $\text{Cu}(\text{UO}_2)_2(\text{PO}_4)_2 \cdot 8\text{--}12\text{H}_2\text{O}$. The poor quality of the available crystals of zeunerite from Schneeberg probably was responsible for the deviation in the later morphological measurements of Schrauf (1872) from those of Weisbach. Schrauf's elements, which correspond in unit and orientation to the x-ray cell found by the present author, are $a:c=1:1.250$. An angle table based on his elements is given in Table 1. Crystals from Schneeberg, described by Ježek (1922) as zeunerite, were probably torbernite; the figures he reported for indices of refraction (Table 2) and specific gravity (3.28) are lower than those found by others for zeunerite.

An x-ray study by the Weissenberg method (copper radiation) was made of zeunerite crystals from the Centennial Eureka mine, Tintic, Utah. The mineral occurs as tiny tablets associated with secondary copper arsenates in oxidized limonitic vein material. The forms $c\{001\}$, $m\{110\}$, $p\{021\}$, $s\{023\}$, and $v\{111\}$ were identified on the crystals by goniometric measurements; the poor quality of the reflections did not

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Contribution from the Department of Mineralogy and Petrography, Harvard University, No. 325.

warrant the derivation of new morphological elements. Rotation, zero-layer, and second-layer photographs were taken about the c axis, and rotation and zero-layer photographs were taken about the two possible choices of horizontal axes. The space group was determined as $P_4/nmm = D_{4h}^7$, it being assumed that the point symmetry is tetragonal holohedral. No x -ray evidence was found for less than tetragonal symmetry. The unit cell dimensions are $a_0 = 7.13 \text{ \AA}$, $c_0 = 8.83 \text{ \AA}$, with $a_0:c_0 = 1:1.238$. The unit cell contents are $\text{Cu}(\text{UO}_2)_2(\text{AsO}_4)_2 \cdot 8\text{H}_2\text{O}$. Beintema (1938) in his work on the structure of meta-autunite I, suggests that only half of the holes in the structure into which the Ca atoms

TABLE 1. ANGLE TABLE FOR NATURAL ZEUNERITE

$$a:c = 1:1.250; p_0:r_0 = 1.250:1$$

Forms	ϕ	ρ	A	\bar{M}
$o \ 001$	—	$0^\circ 00'$	$90^\circ 00'$	$90^\circ 00'$
$a \ 010$	$0^\circ 00'$	$90 \ 00$	$90 \ 00$	$45 \ 00$
$n \ 013$	$0 \ 00$	$22 \ 37$	$90 \ 00$	$74 \ 13$
$f \ 043$	$0 \ 00$	$59 \ 02$	$90 \ 00$	$52 \ 40\frac{1}{2}$
$p \ 021$	$0 \ 00$	$68 \ 12$	$90 \ 00$	$48 \ 58$
$z \ 041$	$0 \ 00$	$78 \ 41\frac{1}{2}$	$90 \ 00$	$46 \ 06$

Doubtful: $m \ 110$, $g \ 012$, $s \ 023$, $y \ 011$, $v \ 111$.

can be fitted are actually occupied by Ca atoms. This explanation may also serve here and account for the fact that the chemical formula gives only one Cu atom per unit cell although the space group demands that there are two equivalent positions for the Cu atom in each unit cell. The calculated specific gravity for the composition is 3.79. The specific gravity of Schneeberg material was redetermined as 3.64. This is probably low, as the crystals contained minute air holes and some inclusions of limonite and the available sample was too small to give an accurate result. The true specific gravity is probably nearer to the calculated value than to that obtained by this measurement.

The x -ray powder pattern data of the Tintic material is given in Table 3. Zeunerite from Schneeberg yielded identical patterns. The presence of arsenic in all the specimens examined was verified by qualitative tests. The indices of refraction, newly measured on this material together with the previously reported measurements, are summarized in Table 2. The observed variation in the indices of refraction suggests that the composition of zeunerite varies in some respects. Possibly there is isomorphous substitution of (PO_4) for (AsO_4) causing a reduction in the

indices of refraction toward those of torbernite; no analytical work has been done to determine whether this is so.

Hallimond (1916, 1920) has shown that torbernite forms three different hydrates: torbernite, meta-torbernite I, and meta-torbernite II. Beintema's work (1938) gave similar information for the isostructural mineral autunite. The water content of each hydrate is in part zeolitic and variable between limits depending on the humidity and temperature. The present study of synthetic zeunerite has shown the existence of at

TABLE 2. OPTICAL DATA FOR NATURAL AND SYNTHETIC ZEUNERITE

All uniaxial negative (—)

	<i>n</i> O	<i>n</i> E
Natural zeunerite		
From Schneeberg*	1.647	—
From Schneeberg*	1.647	—
From Schneeberg*	1.642	—
From Schneeberg*	1.640	—
From Eureka mine, Tintic, Utah*	1.647	1.630
From Eureka mine, Tintic, Utah*	1.643	—
Majuba Hill, Nevada*	1.645	—
Hintze (1931)	1.651	1.635
Millosevich (1912)	1.629	—
Ježek (1922)	1.585	1.576 probably torbernite
Larsen and Berman (1934)	1.643	1.623
Synthetic zeunerite		
With 16H ₂ O	1.602	
With 10H ₂ O	1.610	
With 8H ₂ O	1.645–1.648	
With 5H ₂ O	1.654	

* In the Mineralogical Museum, Harvard University.

least two, and possibly three, hydrates of the copper uranium arsenate. The synthetic material was prepared according to Winkler's (1873) method: freshly precipitated copper carbonate was dissolved in excess arsenic acid, and to this solution uranium nitrate was added, which precipitated the hydrous copper uranium arsenate in distinct but small crystals, less than 0.01 mm. in diameter. Two Penfield water determinations on the fully hydrated material gave 16 molecules of water. After standing in air overnight, the material had dehydrated to 10H₂O and apparently had reached equilibrium. Heating in an oven caused no further loss of water until 65° C was reached. At this temperature the material dehydrated to 8H₂O and stayed in equilibrium up to 85° C.

TABLE 3. INTERPLANAR SPACINGS FROM X-RAY POWDER PATTERNS (Å)
 Cu/Ni radiation = 1.5418 Å.

Synthetic zeunerite				Natural zeunerite	
Fully hydrated (16H ₂ O)		Dehydrated to 8H ₂ O		8H ₂ O	
<i>d</i>	<i>I</i> *	<i>d</i>	<i>I</i> *	<i>d</i>	<i>I</i> *
10.65	10	8.76	10	8.93	10
6.86	5	—	—	6.56	1
5.04	8	—	—	5.95	1
4.55	2	5.47	5	5.54	6
3.59	9	5.04	4	5.04	5
3.39	7	4.35	3	4.35	3
3.18	2	—	—	4.15	1
3.07	1	3.71	9	3.72	9
2.93	4	3.55	7	3.56	7
2.86	4	3.28	8	3.30	8
2.73	3	—	—	3.12	1
2.54	4	2.99	3	3.01	3
2.47	4	2.75	2	2.77	2
2.42	1	2.67	2	2.71	2
2.38	1	2.57	4	2.59	3
2.31	2	2.51	4	2.52	4
2.27	3	2.42	3	2.42	3
2.19	4	2.29	1	2.30	1
2.08	5	2.24	2	2.26	2
1.985	1	2.18	2	2.19	2
1.926	6	2.15	2	2.15	2
1.873	2	2.08	3	2.09	4
1.797	5	1.994	3	2.01	4
1.768	1	1.922	1	1.937	1
1.734	1	1.841	1	1.855	2
1.704	2	1.784	3	1.797	3
1.658	1	1.746	1	1.749	2
1.642	4	1.689	1	1.695	2
1.610	3	1.637	2	1.656	4
1.590	1	1.600	2	1.605	3
1.563	2	1.566	6	1.570	5
1.533	4	1.486	1	—	—
1.512	1	1.451	1	—	—
1.429	3	1.423	1	1.429	2
1.382	3	1.400	1	1.406	2
1.365	3	1.375	2	1.382	3
1.332	1	1.354	1	1.358	2
1.313	2	1.324	1	1.329	2
1.298	1	1.281	1	1.290	1
1.275	3	1.262	1	1.265	1
1.248	2	1.242	1	1.247	1
1.222	1	1.224	1	1.225	1
1.211	2	1.208	1	1.209	1
1.202	2	1.175	2	1.176	2
1.191	1	1.160	1	1.161	1
1.181	1	1.127	2	1.130	1
1.168	2	1.118	1	—	—
1.143	2	1.088	1	1.092	1
1.118	1	1.077	1	—	—
1.094	2	1.065	1	1.055	1
1.086	2				
1.060	1				

* Intensities estimated visually.

From this temperature up to 105°C the material lost water slowly but continuously and finally reached equilibrium with $5\text{H}_2\text{O}$. This hydrate, however, is not stable under atmospheric conditions; after standing in air for one day it had rehydrated to $8\text{H}_2\text{O}$. The entire dehydration process, from 16 to 5 molecules of water, took about two weeks.

Bergman (1925) dehydrated zeunerite over sulfuric acid and changed the vapor pressure by changing the concentration of the acid. He indicated the existence of at least two hydrates. From Fig. 1, which graphically represents the course of dehydration with increase in temperature, it would appear that there are four hydrates of the synthetic material. X-ray powder patterns for the material with 16 molecules of water and 10 molecules of water are the same. Also, the x-ray powder patterns (Fig. 2) for the hydrates with 8 and 5 molecules of water are

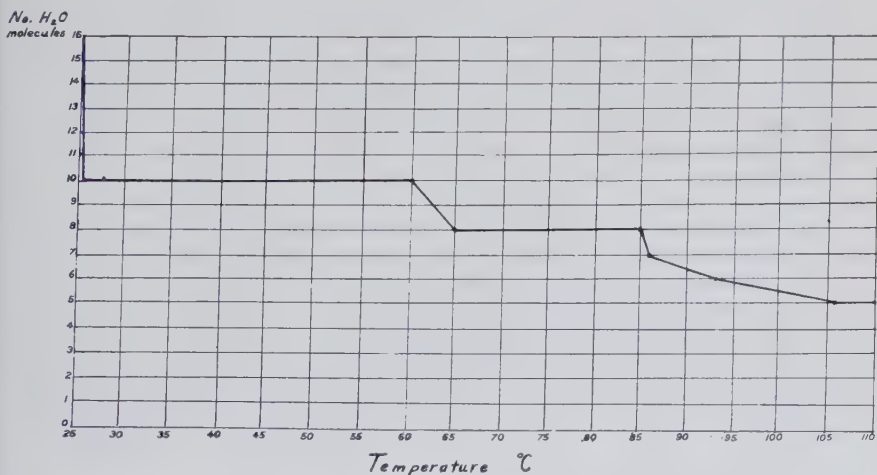


FIG. 1. Dehydration Curve for Synthetic Zeunerite.

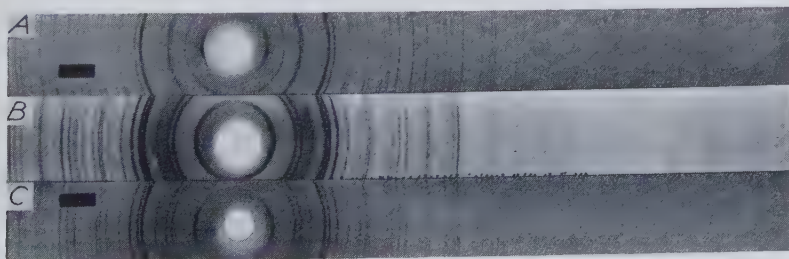


FIG. 2. X-ray Powder Patterns.

- A. Synthetic zeunerite, fully hydrated, $16\text{H}_2\text{O}$.
- B. Synthetic zeunerite, dehydrated to $8\text{H}_2\text{O}$.
- C. Natural zeunerite, with $8\text{H}_2\text{O}$.

identical and are similar to the patterns of natural zeunerite, meta-autunite I, and meta-torbernite I. One sample of dehydrated material with $5\text{H}_2\text{O}$ was allowed to remain in the oven for 24 hours at 110°C . This material gave an x -ray powder pattern different from the others but like those of meta-autunite II and meta-torbernite II. The pattern was faint, and, although attempts were made to obtain a better one, the results could not be duplicated. All other samples with $5\text{H}_2\text{O}$ gave patterns like those of the material with $8\text{H}_2\text{O}$. It is possible that above 110°C the material will dehydrate still further, perhaps down to $2\frac{1}{2}$ molecules of water, like meta-autunite II. The copper uranium arsenate thus may occur as three hydrates, each with a water content that varies within certain limits according to temperature and humidity. Beintema found that the change from autunite to meta-autunite I was reversible under suitable atmospheric conditions. The behavior of zeunerite is somewhat different. The synthetic material with $8\text{H}_2\text{O}$ (corresponding to meta-autunite I) apparently is stable, even in a humid atmosphere (Cambridge, Massachusetts, in the summer), and natural zeunerite with $8\text{H}_2\text{O}$ failed to hydrate further after soaking in cold water for seven days.

Variation in water content of zeunerite causes a variation in the indices of refraction. The index of refraction (n_O) of natural zeunerite varies from 1.629 to 1.651 (see Table 2). The fully hydrated synthetic material has $n_O = 1.602$. With a decrease in water content to $10\text{H}_2\text{O}$, n_O increases to 1.610. The considerable increase in n_O to 1.645–1.648 with the loss of only two more molecules of water is consistent with the x -ray data which show a change in crystal structure at this point in the dehydration process (Table 3). Further dehydration to $5\text{H}_2\text{O}$ caused an increase of n_O to 1.654. Upon standing in air the synthetic material with $5\text{H}_2\text{O}$ rehydrated to $8\text{H}_2\text{O}$ and n_O lowered to 1.645.

Fluorescence also varies with water content. All of the natural zeunerite specimens fluoresced weakly under both long- and short-wave ultraviolet light. The fully hydrated synthetic zeunerite did not fluoresce; the air-dried material ($10\text{H}_2\text{O}$) showed faint yellow-green fluorescence under long-wave ultraviolet light; and the heated material dehydrated to $8\text{H}_2\text{O}$ fluoresced faintly yellow-green under the short-wave ultraviolet light and somewhat more strongly under the long-wave ultraviolet light. The property of fluorescence in these minerals, then, is not dependent solely upon the nature of the divalent cations, as suggested by Meixner (1940), but also upon the number of water molecules in the structure. A similar relationship of fluorescence to degree of hydration has been noted by Axelrod et al. (1951) for the minerals bayleyite and andersonite.

To be consistent with the nomenclature used in the torbernite and

autunite studies, the natural mineral zeunerite might be called meta-zeunerite, and the possibility that a higher hydrate (true zeunerite) exists in nature as it does in the synthetic material must be considered. One hesitates, however, to set aside an established name and to assign a new name in anticipation of finding another mineral species, no matter how well grounded the reasoning behind this assignment may be.

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MINERALOGY OF TRIPLITE

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ABSTRACT

Triplite, previously treated as a divariant series between Fe" and Mn", may also contain relatively large amounts of Mg and lesser amounts of Ca. Variation in Fe and Mg is the primary factor influencing the optical properties, but Mn variation has little effect. Three new analyses of Colorado triplites are presented; two are high in Mg. Apparently the only valid species formed through weathering of triplite are dufrenite, vivianite, and phosphosiderite. Alluaudite and apatite form by reaction with solutions. Nearly 80 triplite localities have been recorded. Triplite occurs in four types of granitic pegmatites and three types of hydrothermal, high temperature veins.

INTRODUCTION

Previous to the work of Hurlbut (1936) triplite generally was regarded as consisting of an isomorphous series of two elements, ferrous iron and manganese. Analyses of triplite containing important amounts of magnesium were rare, and usually no consideration was given to the element in attempting to relate physical properties to composition (Henderson, 1928, Otto, 1936, and Richmond, 1940). As a result of a study of the pegmatites of Eight Mile Park, Fremont County, Colorado, the writer described several new triplite discoveries (Heinrich, 1948). Rough crystals of triplite from one of these occurrences were measured by Wolfe and Heinrich (1947). Qualitative tests indicated that the magnesium contents of these triplites might be abnormally large and that complete chemical analyses would be desirable. The writer is indebted to the Faculty Research Fund of the Rackham Graduate School, University of Michigan, for a generous grant defraying the cost of the analyses and other expenses. Thanks are due to Professor Clifford Frondel of the Department of Mineralogy and Petrography, Harvard University, and to Professor Brian Mason of the Department of Geology, Indiana University, for critical examinations of the manuscript and for suggestions toward its improvement. All conclusions are the responsibility of the writer. Dr. George Switzer of the U. S. National Museum kindly furnished several specimens for examination, and Mr. W. G. Griffiths of the U. S. Geological Survey furnished a description of and a specimen from a previously unrecorded occurrence in northern Colorado.

NEW ANALYSES

In Table 1 are listed the results of three new analyses of triplite. All samples were taken from unaltered central parts of triplite masses that did not contain any other intergrown minerals. Selected pieces were

broken to minus 1/8th-inch particles and handpicked under a binocular microscope. Pieces showing alteration or coatings were rejected, and the remainder was ground in an agate mortar. The powdered sample was

TABLE 1. NEW ANALYSES OF TRIPLITE

	1.	1a	1b	2.	2a	2b	3.	3a	3b
Fe ₂ O ₃	0.86			1.63			2.30		
TiO ₂	0.19			0.08			0.42		
FeO	9.00	.125		10.65	.168*		11.39	.187*	
MnO	33.20	.468		44.44	.626		31.46	.443	
MgO	13.62	.337	4	1.15	.028	4	9.93	.246	4
CaO	2.29	.040		4.59	.082		4.70	.083	
Na ₂ O	0.03			0.11			0.11		
K ₂ O	0.01			0.03			0.02		
H ₂ O+	0.21			0.23			0.35		
H ₂ O—	0.03			0.06			0.06		
P ₂ O ₅	34.96	.246	1	32.01	.225	1	34.41	.242	1
F	8.52	.448	2	7.66	.403	2	7.63	4.01	2
Total	102.92			102.64			102.78		
Less O for F	3.59			3.25			3.22		
Total	99.33			99.39			99.56		

1. Triplite, Mica Lodge pegmatite, Eight Mile Park, Fremont County, Colorado.

1a. Molecular ratios.

1b. Molecular proportions.

2. Triplite, School Section pegmatite, Eight Mile Park, Fremont County, Colorado.

2a. Molecular ratios.

2b. Molecular proportions.

3. Triplite, Turkey Creek, Deadmans Canyon, El Paso County, Colorado.

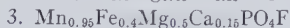
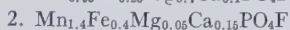
3a. Molecular ratios.

3b. Molecular proportions.

Analyst, Lee C. Peck.

* With Fe₂O₃ recalculated to equivalent FeO.

The formulae are:



examined in oil immersion and was found to be practically free of any impurities. Analyses of three-gram samples were completed at the Laboratory for Rock Analysis, Department of Geology and Mineralogy, University of Minnesota, by Dr. Lee C. Peck, to whom the writer is strongly indebted for painstaking work in difficult analyses.

Triplite from the Mica Lode (Anal. No. 1) has one of the highest MgO contents reported for the mineral, being exceeded only in the material from Punau, Marienbad, Moravia (Anal. No. 27, Table 2) and "talctriplite" from Horrsjöberg, Sweden (Anal. No. 30, Table 2.) Both of these older analyses (1910 and 1883, respectively) are of poorer quality.

4. Branchville, Conn. Dana, 1892, p. 777, Anal.—Penfield.
5. Haddam Neck, Conn. Shannon, 1920. Anal.—Shannon. Rem. is insol.
6. Rapid City, South Dakota. Eakins, 1891. Anal.—Eakins. Rem. is SiO_2 0.43, Al_2O_3 8.74, K_2O tr, Na_2O 5.25, Li_2O 0.13, Cl 0.25, CO_2 0.26. "In reality it probably represents a mixture . . ."
7. Reagan Mining District, Aurum, White Pine County, Nevada. Hess and Hunt, 1913. Anal.—Hunt.
8. 7U7 Ranch, near Hillside, Arizona. Hurlbut, 1936. Anal.—Gonyer. Rem. is Na_2O .
9. Mt. Loma, 7 miles from locality of No. 8, near Hillside, Arizona. Hurlbut, 1936. Anal.—Gonyer.
10. San Luis, Sierrá de Córdoba, Argentina. Stelzner, 1873. Anal.—Siewart. Rem. is SiO_2 .
11. San Luis, Sierrá de Córdoba, Argentina. Stelzner, 1873. Anal.—Siewart. Rem. is SiO_2 .
12. Salado, La Rioja, Argentina. Henderson, 1933. Anal.—Henderson. Rem. is Na_2O 0.27, K_2O 0.33, insol. 0.28, Cl tr.
13. Mangualde, Portugal. de Jesus, 1933. Anal.—de Jesus. Rem. is Na_2O 0.38, K_2O 0.27, $-\text{H}_2\text{O}$ 0.07, insol. 0.24.
14. Mangualde, Portugal. de Jesus, 1933. Anal.—de Jesus. Rem. is Na_2O 0.82, K_2O 1.18, $-\text{H}_2\text{O}$ 2.50, insol. 0.20, Mn_2O_3 4.74. Called metatriplite.
15. Chanteloube, Limoges, France. Otto, 1936. Anal.—Otto. Fluorine not determined. Two older analyses of triplite from France are not included in the table; both are of material from Limoges (Vauquelin, 1802 and Berzelius, 1819.) Vauquelin presented the original description of triplite, under the name, "phosphate native de fer mélange de manganèse." The name, triplite, was applied by Hausmann in 1813.
16. Zwiesel, Bavaria. Fuchs, 1839. Anal.—Fuchs. Rem. is SiO_2 0.68, Fe 4.76.
17. Zwiesel, Bavaria. Otto, 1936. Anal.—Otto.
18. Schönfeld, Germany. Otto, 1936. Anal.—Otto.
19. Wildenau-Plössberg, Germany. Laubman and Steinmetz, 1920. Anal.—authors. Note low P_2O_5 . Rem. is insol.
20. Pleystein, Germany. Laubman and Steinmetz, 1920. Anal.—authors. Rem. is insol.
21. Hagendorf, Germany. Laubman and Steinmetz, 1920. Anal.—authors. Rem. is insol.
22. Schlaggenwald, Bohemia. von Kobell, 1864. Anal.—von Kobell.
23. Vienna, Gross-Meseritsch, Moravia. von John, 1900. Anal.—von John. Rem. is Na_2O 0.52, K_2O 0.72.
24. Vienna, Gross-Meseritsch, Moravia. Kovar and Slavik, 1900. Anal.—Kovar. Rem. is SiO_2 . Altered, may be triplodite.
25. Königswart, Marienbad, Moravia. Sellner, 1924. Anal.—Stanczak. Rem. is Na_2O 0.22, K_2O 0.06, insol. 0.36.
26. Königswart, Marienbad, Moravia. Sellner, 1924. Anal.—Stanczak. Rem. is Na_2O 0.30, K_2O 0.12.
27. Punau, Marienbad, Moravia. Sellner, 1924. Anal.—Stanczak. Rem. is Na_2O 0.36, K_2O 0.45, insol. 8.10. Probably impure, note high insol. " . . . gangformig Triplit mit Granat." According to Lazaravicz (1910) also intergrown with fluorite.
28. Peilau, Reichenberg, Silesia. Bergemann, 1860. Anal.—Bergemann. Rem. is SiO_2 0.23, Na_2O 0.41, ignition loss 1.28. Fluorine not determined.
29. Varuträsk, Sweden. Mason, 1941B. Anal.—Berggren. Rem. is Na_2O 0.39, K_2O 0.33, Li_2O 0.10, $-\text{H}_2\text{O}$ 0.27, Insol. 6.22.
30. Horrsjöberg, Sweden. Igelström, 1882. Anal.—Igelström. Fluorine not determined. Very likely a mixture, for Igelström (1883) states, " . . . je n'avais alors que peu d'échantillons assez purs."
31. Skrupetorpsee, Östergötland, Sweden. Hamberg, 1904. Anal.—Sahlbohm. Rem. is SiO_2 0.11, Al_2O_3 2.16, Na_2O 0.98, K_2O 0.33.
32. Lilla Elysjö, Krockek, Linköping, Sweden. Nordenskjöld, 1902. Anal.—Nordenskjöld. Rem. is SiO_2 0.18, Al_2O_3 0.37, Na_2O 0.31.
33. Lemnäs, Kimito, Southwest Finland. Pehrman, 1939. Anal.—Pehrman. Rem. is Na_2O 0.22, K_2O tr, $-\text{H}_2\text{O}$ 0.07.
34. Mattkärr, Kimito, Finland. Otto, 1936. Anal.—Otto.
35. Serro Branco, Picubhy, Parahyba, Brazil. Fornaseri, 1943. Anal.—Fornaseri. Rem. is Na_2O 0.10, K_2O tr, $-\text{H}_2\text{O}$ 0.20. Called "alluaudite."

TABLE 2. OTHER ANALYSES OF TRIPLITE

No.	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
FeO	7.69	4.95	1.97	1.88	11.68	6.68	18.30	15.88	23.54	20.34	—	27.61	35.44	41.96	25.80	41.60
Fe ₂ O ₃	—	—	2.36	—	—	0.40	—	2.22	—	2.50	24.20	—	—	—	—	—
MnO	54.14	52.40	29.13	57.63	34.55	53.77	37.84	37.74	34.84	36.52	29.90	33.44	20.34	18.40	28.48	31.03
MgO	—	0.58	tr.	1.21	11.87	0.31	tr.	tr.	tr.	0.91	0.17	—	—	0.80	7.20	—
CaO	1.80	3.18	6.72	2.86	2.48	2.17	4.46	5.92	3.48	2.86	2.18	1.70	—	1.69	1.80	—
H ₂ O+	0.36	0.35	3.67	—	0.75	—	—	—	1.58	0.13	4.70	—	—	—	—	—
P ₂ O ₅	32.17	32.81	39.68	31.84	33.32	32.20	35.65	31.13	31.50	32.15	26.67	32.02	35.60	32.85	31.98	19.73
F	7.53	9.09	2.35	7.77	8.02	7.58	4.94	7.78	6.41	7.35	5.02	n.d.	3.18	7.21	7.64	6.28
Rem.	—	1.17	15.06	—	0.52	—	0.13	1.17	0.88	0.96	9.44	—	5.44	—	—	1.03
Total	103.69	104.53	100.94	102.99	103.19	103.11	101.32	101.84	102.23	103.72	102.28	94.77	100.00	102.91	102.90	99.67
O=F	3.16	3.83	1.05	3.27	3.38	3.19	2.07	3.27	2.69	3.09	2.11	—	1.37	3.02	3.21	2.64
Sum	100.53	100.70	99.89	99.72	99.81	99.92	99.25	98.57	99.54	100.63	100.17	94.77	98.63	99.89	99.69	97.03

No.	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35
FeO	33.51	35.50	26.98	31.03	26.10	24.84	26.50	14.87	31.72	19.96	16.12	20.48	18.43	21.90	38.52	29.40
Fe ₂ O ₃	—	—	—	—	2.80	0.17	1.43	—	1.55	0.78	—	3.40	2.38	—	—	0.10
MnO	25.42	28.66	30.00	31.05	29.17	31.96	33.40	20.75	30.83	32.07	14.86	32.60	35.23	37.35	24.20	25.97
MgO	—	—	3.05	tr.	4.58	tr.	tr.	14.05	0.32	tr.	17.42	1.33	4.46	—	—	1.60
CaO	—	—	2.20	1.42	0.49	6.89	4.90	7.84	1.19	4.68	14.91	0.80	2.10	2.65	0.50	8.10
H ₂ O+	—	—	—	—	4.16	—	—	—	—	0.48	—	1.25	0.10	0.38	0.62	1.02
P ₂ O ₅	33.14	31.89	33.85	31.29	31.67	31.87	30.44	29.08	32.76	30.63	32.82	32.33	32.05	33.09	32.65	31.96
F	6.77	7.29	8.10	8.17	1.11	3.29	3.34	5.15	n.d.	6.72	n.d.	6.96	8.72	7.83	6.02	5.50
Rem.	0.46	0.83	—	1.24	0.84	0.64	0.42	8.91	1.92	7.31	—	3.58	0.86	0.29	—	0.30
Total	99.30	104.17	104.18	104.20	100.92	99.66	100.43	100.65	100.29	102.63	96.13	102.73	104.33	103.49	102.51	102.95
O=F	2.84	3.06	3.40	3.43	0.47	1.39	1.40	2.16	—	2.83	—	2.84	3.66	3.29	2.57	2.31
Sum	96.46	101.11	100.78	100.77	100.45	98.27	99.03	98.49	100.29	99.80	96.13	99.89	100.67	100.20	99.94	100.64

See opposite page for notes.

OTHER ANALYSES

Table 2 lists analyses of triplite from the literature. Those in which fluorine was not determined are of questionable value, and those that list a high percentage of insoluble material were undoubtedly performed on impure samples. Large percentages of Fe_2O_3 , Mn_2O_3 , and $\text{H}_2\text{O}(-)$ and subnormal percentages of P_2O_5 and F indicate alteration of the specimen.

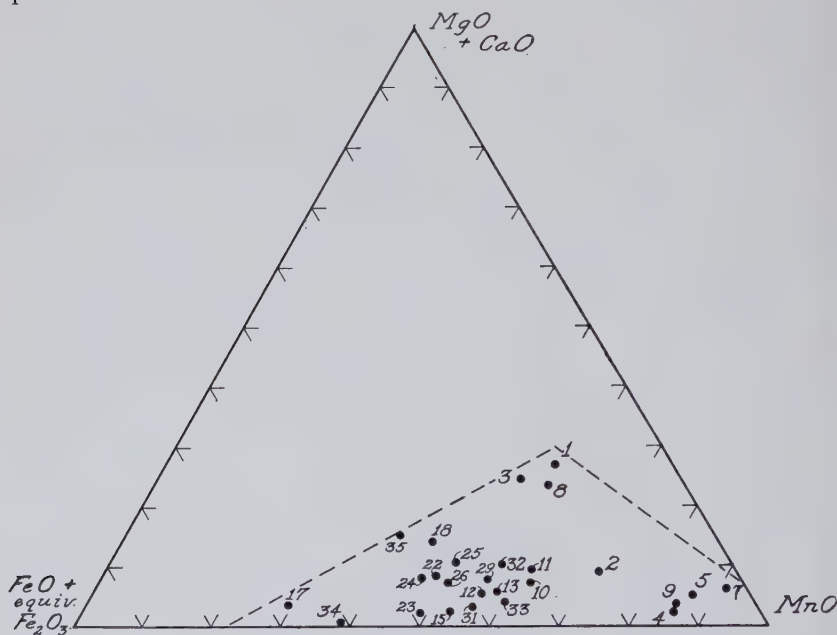


FIG. 1. Compositional field of triplite in terms of $\text{FeO} + \text{equivalent } \text{Fe}_2\text{O}_3$, MnO , and $\text{MgO} + \text{CaO}$.

Analyses of good quality, i.e., those with low Fe_2O_3 , low $\text{H}_2\text{O} +$, high P_2O_5 , and low insoluble material, are plotted on the triangular diagram of Fig. 1, whose corners are MnO , $\text{FeO} + \text{equivalent } \text{Fe}_2\text{O}_3$, and $\text{MgO} + \text{CaO}$, recalculated to 100%. The dashed lines define the compositional field of triplite. The maximum iron recorded is 41.96% FeO (Otto, 1936) in triplite from Zwiesel, Bavaria (Anal. 17). The MnO maximum is 57.63% in material from Nevada (Hess and Hunt, 1913, Anal. 7). The highest MgO , 13.62%, in a modern analysis is from the Mica Lode triplite (Anal. 1). Analyses 27 and 30 show higher MgO contents, but both are of doubtful quality. This last analysis also has an abnormal CaO content above that of Anal. 35, which is apparently a good-quality analysis of triplite from Brazil and which contains 8.10% CaO .

Triplite does not form an isomorphous series with wagnerite. Intermediate compounds are unknown, and b_0 of wagnerite is approximately twice that of triplite. Neither does triplite form an isomorphous series with triploidite, which was recognized by Frondel (1949). Most triplites with less than about 7% F also show some oxidation of the iron.

RELATION OF PHYSICAL PROPERTIES TO COMPOSITION

Indices of refraction

Indices of refraction have been measured on only 14 analyzed specimens (Table 3).

TABLE 3. INDICES OF REFRACTION OF ANALYZED TRIPLITES

Anal No.	1	2	3	5	7	8	9
α	1.643	1.671	1.648	1.665	1.650	1.651	1.662
β	1.647	1.681	1.652	1.673	1.660	1.653	1.673
γ	1.668	1.686	1.672	1.682	1.672	1.665	1.684

Anal No.	12	15	17	18	29	33	34
α	1.675	1.680	1.696	1.664	1.673	1.677	1.684
β	1.683	1.686	1.704	1.674	1.681	1.685	1.693
γ	1.692	1.695	1.713	1.680	1.691	1.695	1.703

Otto (1936) treated triplite as a two-variable series between Fe'' and Mn'' and failed to consider the modifying influence of Mg and Ca on the indices of refraction. His graph (p. 95) shows a straight line relation between % FeO and the three indices.

With those analyses of Fig. 1 for which indices are available, it is possible to contour the triangular diagram for values of the refractive indices. This has been done for α (Fig. 2) and γ (Fig. 3). The few discrepancies that appear probably are attributable to some inaccuracies in index determination. For example, in Fig. 2, α of Anal. 9 appears to be too low and in Fig. 3, γ of Anal. 8 is apparently low.

The contour patterns clearly reveal: (1) that the indices increase with increasing FeO; (2) that the indices decrease with increasing $\text{MgO} + \text{CaO}$; (3) that the rate of increase per unit of FeO is much greater than the rate of decrease per unit of $\text{MgO} + \text{CaO}$; (4) that variations in the MnO

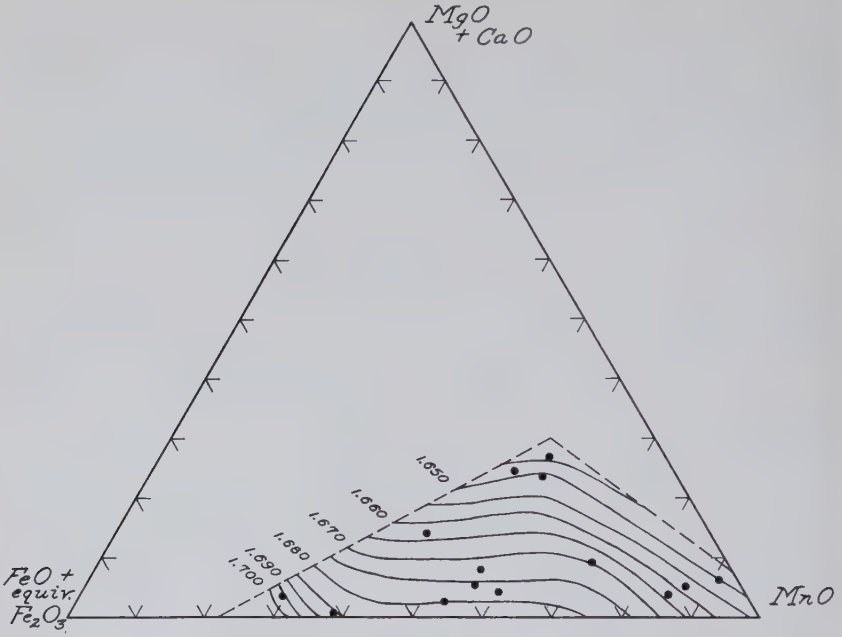


FIG. 2. Relationship between composition and α index of triplite.

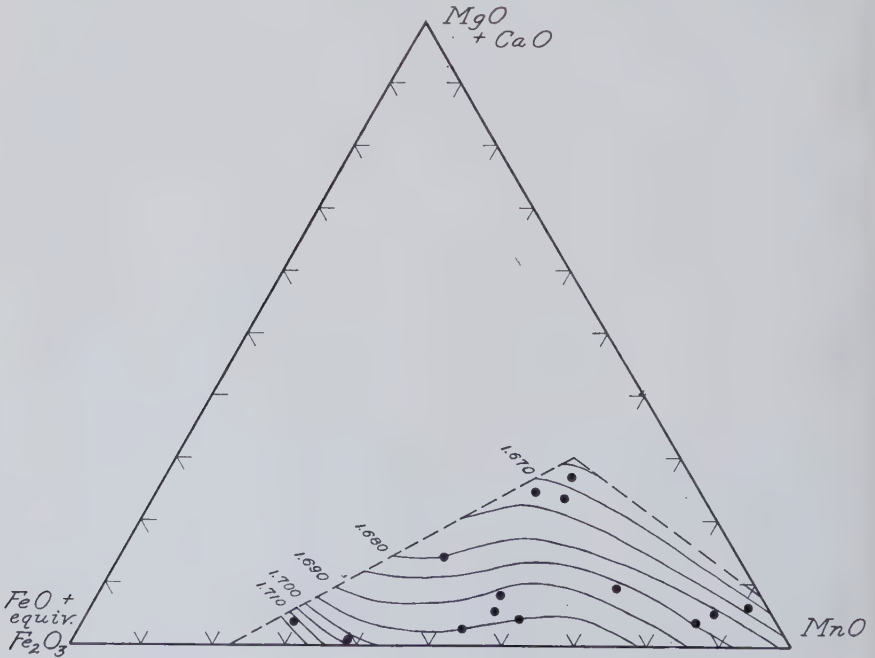


FIG. 3. Relationship between composition and γ index of triplite.

content have almost negligible effect upon changes in the values of the indices.

If triplite contains very minor amounts of $\text{MgO} + \text{CaO}$, it may be treated as a two-variable system in which the indices of refraction change directly with the iron content. This relationship is shown graphically in Fig. 4, in which γ is plotted against $\frac{\text{FeO} + \text{equiv. Fe}_2\text{O}_3}{\text{MnO}}$, for four

triplites with $\text{MgO} + \text{CaO}$ less than 3.00%. However, relatively few triplites fulfill this last condition.

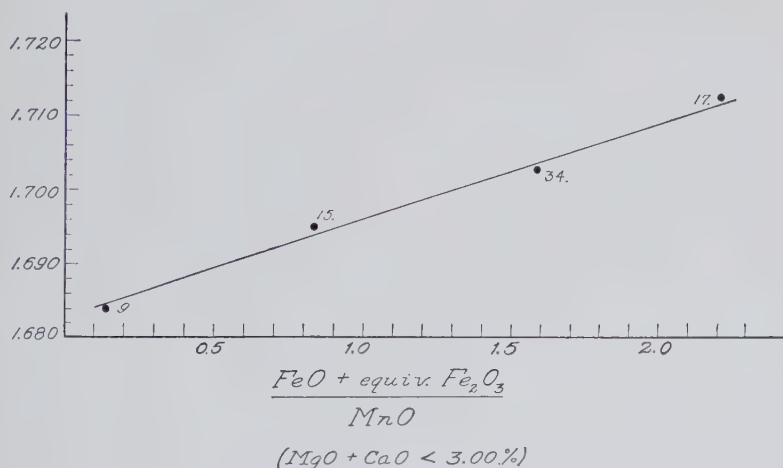


FIG. 4. Relationship between γ index and $\frac{\text{FeO} + \text{equiv. Fe}_2\text{O}_3}{\text{MnO}}$ in triplites that have $\text{MgO} + \text{CaO} < 3.00\%$.

Henderson (1928) employed the name *zwieselite* for iron-rich triplite. However, since three variables and not two are important in triplite, this designation should not be retained and the varietal name *ferroan triplite* is preferable.

Other Optical Properties

The optical orientation of triplite varies with the composition, although too few data are available to systematize the relationships. It seems clear that with increasing amounts of MgO and CaO the optical angle ($2V$) decreases to as low as 25° , for $\text{MgO} + \text{CaO} = 15.91\%$ (Anal. 1). Most triplites are optically positive with a large $2V$ and $b = Y$. In two cases, however, the mineral is optically negative with a large $2V$ (Anal. 2 and 9). Figures 5a and 5b illustrate the differences in optical

orientation for two triplite of markedly different chemical composition.

Hurlbut (1936) reported a peculiar orientation for triplite from the 7U7 Ranch, Arizona, which was said to have $X=b$, $Z \wedge a = 22^\circ$, $2V = 28^\circ$, and to be optically positive. This orientation was based on the assumption that (100) was the best cleavage (Hurlbut, priv. comm.). More recent work indicates that (001) is the best cleavage with (010) as next

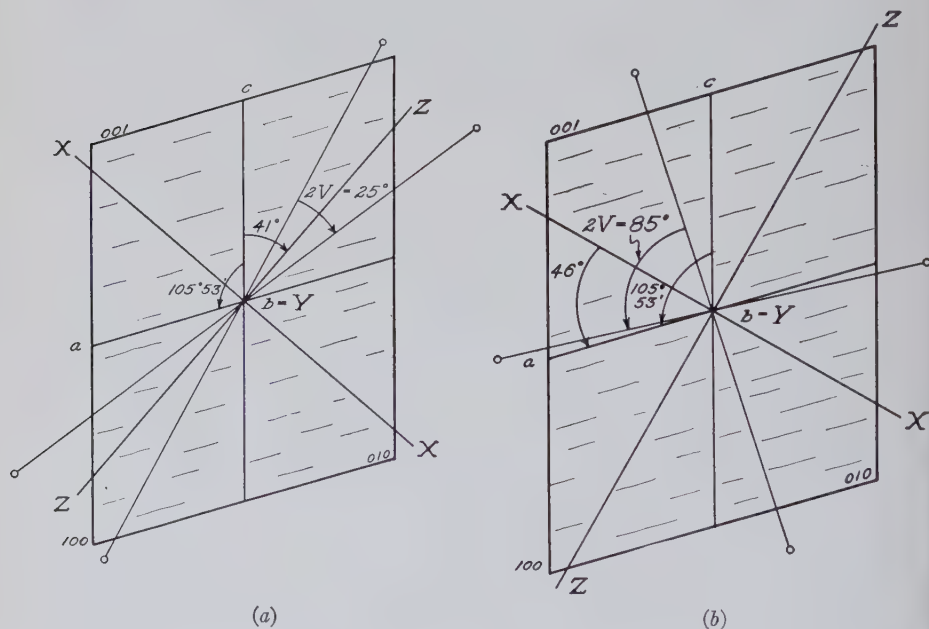


FIG. 5. Optical orientation of triplite. *a.* From Mica Lode (Anal. 1);
b. From School Section (Anal. 2).

best (Pehrman, 1939, Richmond, 1940, and Wolfe and Heinrich, 1947) and (301) as another possible cleavage (Pehrman, 1939). The front pinacoid (100) was found by Wolfe and Heinrich (1947) as a poor cleavage direction on only one crystal from the Mica Lode pegmatite. If the new cleavage orientation is applied to Hurlbut's data and it is assumed the cleavage he used as (100) is actually (010), his optical orientation becomes inconsistent with a monoclinic symmetry. Unfortunately, it was impossible to check this orientation, for a specimen of triplite from this locality kindly submitted to the writer from the Harvard Mineralogical Museum proved to be badly weathered and nearly opaque due to secondary oxides of iron and manganese. Type material from Mt. Loma, Arizona (Anal. 9), also described by Hurlbut (1936) was checked by the

writer and found to have an optical orientation very similar to that for School Section material (Anal. 2) (Fig. 5b).

Triplite shows considerable variation in color from salmon pink to dark brown with the darker colors prevailing in the iron-rich varieties and the lighter colors more characteristic for types poorer in iron. Variable amounts of magnesium and calcium have apparently no marked effect on the color. The absorption formula is variable; recorded examples are: Lemnäs, Kimito, Finland, $X < Y < Z$ (Anal. 33); La Rioja, Argentina, $X > Y > Z$ (Anal. 12); Reagan Mining District, Nevada, $X > Y \approx Z$ (Anal. 7); Haddam Neck, Conn., $X > Z > Y$ (Anal. 5); and Mica Lode, Colo., $X = Z > Y$ (Anal. 1).

Pebrman (1939) found an irregular color distribution in triplite from Lemnäs, Kimito, Finland, and ascribed the origin of the colorless patches to local replacement of iron and manganese by calcium. Similar color irregularities occur in triplite from the Mica Lode pegmatite. Not only do randomly distributed patches show a lower color density, but they also are characterized by a markedly lower dispersion and somewhat lower birefringence.

Henderson (1933) states, "There is a rather prominent zoned structure present and these zones are composed of triplite which has a duller luster and a more pronounced brown color without the reddish tint. When the mineral is powdered and examined under the microscope these different bands are indistinguishable from each other." This zoning may well be that due to incipient alteration so commonly found in triplite. The relative positions of the zones are not given by Henderson. Zoned triplite also has been reported by Glass (1935) from the Morefield mine, Virginia, where cores are salmon colored and outer parts are brown-black. This too may be the result of alteration.

The dispersion is $r > v$ and ranges from weak to strong.

Specific Gravity

There are available 22 specific gravity determinations on analyzed triplite including five by the writer (Anal. 1, 2, 3, 8, and 9). These analyses were plotted on the triangular diagram used as a base for Figs. 1-3. It was not possible, however, to contour the diagram in terms of specific gravity values. This appears to be due, in part, to wide variations in the quality of the determinations and in part to the influence of variations in the fluorine content. The value of G in unaltered triplites ranges from 3.55 to 3.97. In general the specific gravity increases with increasing FeO and decreases with increasing MgO+CaO.

That the variation in fluorine has a marked effect on G is shown in Fig. 6. For three triplites, whose FeO+equivalent Fe_2O_3 , MnO, and

MgO+CaO contents are similar, G is plotted against weight percent fluorine. The graph reveals a significant decline in G with increasing fluorine, other elements being nearly constant.

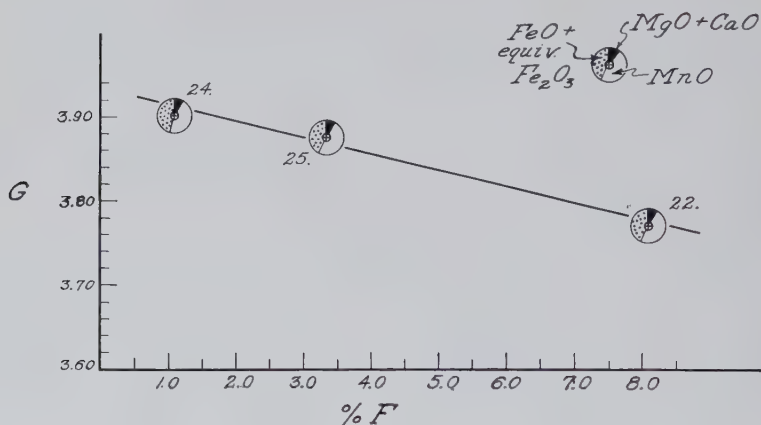


FIG. 6. Relationship between specific gravity and fluorine content in triplites whose contents of FeO, MnO, and MgO+CaO are similar.

Alteration

The following minerals have been classed as alteration products of triplite: pseudotriplite (Blum, 1845), "a substance near heterosite" (Dana, 1892), alluaudite (Dana, 1892 and Hintze, 1933), dufrenite (Kovar and Slavik, 1900), phosphosiderite (Sellner, 1924), heterosite and purpurite (Müllbauer, 1925), metatriplite (de Jesus, 1933), lemnäsite (Pehrman, 1939), apatite (Pehrman, 1939 and Heinrich, 1948), and vivianite (Sellner, 1924 and Mason, 1941B). The alteration products of iron-manganese phosphate minerals have been thoroughly investigated by Mason (1941A) who demonstrates the following:

1. That pseudotriplite was originally described as an alteration of triphyllite by Fuchs (1835) and furthermore is synonymous with heterosite.
2. That alluaudite contains sodium and was considered by Lacroix (1910) to be an alteration of natrophilite.
3. That the heterosite of Müllbauer (1925) is actually ferri-sicklerite and his purpurite is heterosite, and that they both clearly are alterations of triphyllite and not of triplite. Mason (1941A, p. 149) states, "Derivation of heterosite and purpurite from triplite must be considered doubtful, to say the least."

Kovar and Slavik (1900) state that the alteration of triplite begins with replacement of fluorine by hydroxyl to form triplodite and continues with the subsequent oxidation of iron and manganese. They cite as evidence for the first change two analyses of "triplite," one from

Vienna, Moravia, with only a trace of fluorine and the other from Cyrillhof, Moravia, with 0.88% fluorine. These are, of course, triploidite, and not triplite, as was also recognized by Frondel (1949). The authors give no evidence, other than the low fluorine contents, that either was derived from triplite through replacement of fluorine by hydroxyl. In fact they state specifically that the analysis of the Cyrillhof mineral (renamed wolfeite by Frondel, 1949) was conducted on "Möglichst frische Triplitstücke" (p. 401). Thus it appears probable that they were dealing with iron-rich triploidite of primary rather than of secondary origin and that the Vienna pegmatite contained both triplite and triploidite. The coexistence of triplite and triploidite in the same pegmatite is reported from Branchville, Connecticut; Hagendorf, Bavaria; Skrumpetorp, Sweden (wolfeite); and Vavřinec Hill, Bohemia. There is no proof that triploidite forms by alteration of triplite, although Scholz (1925) also states that triplite plus water will yield triploidite.

Mason (1941A) has shown that all of the iron in iron-manganese phosphates will be oxidized to the ferric state before any of the manganese is changed to the manganic state. The oxidation of the iron and replacement of fluorine in triplite probably proceed simultaneously. This is shown by analysis 14 (metatriplite from Portugal), in which all of the iron and some of the manganese have been oxidized, but much of the fluorine remains. The material, metatriplite (de Jesus, 1933), represents an early stage in the alteration of triplite. The structural identity of the type metatriplite with ordinary triplite has been established by Mason (pers. comm.) by means of x-ray powder photographs. Metatriplite very likely is the same material that commonly forms a thin darker, non-vitreous alteration shell around many triplite masses. This material has a lower specific gravity and lower indices than triplite and is nearly opaque due to very abundant minute dark brown inclusions (specks of Fe_2O_3 and Mn_2O_3 ?). It, too, has been shown to be structurally identical with triplite (Wolfe and Heinrich, 1947).

The only valid species formed as weathering products of triplite are, therefore, dufrenite, phosphosiderite, and vivianite. Lemnäsite, which Mason (1941A) has shown to be identical with alluaudite, is said by Pehrman (1939) to have been formed from triplite by reaction with Na-bearing solutions that also formed cleavelandite and is thus of hydrothermal origin. Similarly the dark manganiferous apatite described by the same author as forming shells on triplite in the Lemnäs pegmatite must be a reaction product with Ca solutions. Dark brown apatite found by the writer (Heinrich, 1948) as an alteration of triplite is believed to have been formed through supergene alteration by CaCO_3 waters.

OCCURRENCE AND DISTRIBUTION

Triplite has been found at the following localities:

- U.S.A. Maine:* Auburn; Harndon Hill, Stoneham; Buckfield (a U. S. Nat. Mus. spec. from this locality is apatite). *Connecticut:* Haddam Neck; Middletown; Branchville; Paul Anderson farm, 5 miles south of Cobalt. *New Hampshire:* Palermo Mine, North Groton. *Virginia:* Rutherford and Morefield mines, Amelia. *South Dakota:* Etta Mine, Keystone; High Climb Mine, Custer; Margaret Tin Mine, Pennington Co.; an unidentified locality cited as "Rapid City." *Colorado:* Mica Lode and School Section pegmatites, Eight Mile Park, Fremont Co.; Deadmans Canyon, Turkey Creek, El Paso Co.; Big Boulder pegmatite and two other pegmatites $\frac{1}{2}$ mile to the northeast, Crystal Mountain Area, Larimer Co. *Arizona:* 7U7 Ranch and Mt. Loma, near Hillside; near Wickenburg. *Nevada:* Southeast side Kern Mtns., near Reagan, White Pine Co. *California:* Stewart Mine, Pala; Camp Signal, 9 miles north of Goffs, San Bernardino Co.
- Argentina:* Salado, La Rioja; San Luis and San Rogue, Sierrá de Córdoba; San Judas Tadeo pegmatite, Punilla Valley, Córdoba. *Bolivia:* Near Fabulosa Mine, headwaters of Rio Lallana, Cordillera Real. *Brazil:* Serro Branco, Picuhy, Parahybla.
- Portugal:* Mangualde. *France:* Alluad, La Vilate, and Chatres, all near Chanteloube; Chabanne and Hureaux, in Saint Sylvestre; Compreignac; Bessines; Charmasse, in Mesvre, Saône-et-Loire.
- Germany:* Marchaney, Wildenau-Plössberg, Pleystein, Hühnerkobel, Hagendorf, Döfering near Waldmünchen, Frath, Blötz, Brandten, Harlachberg, Hörnelberg, Birkhöhe and Kammermayer-Keller by Zwiesel, all in Bavaria; Geyer, Saxony; Schönfeld (state unknown).
- Bohemia:* Vavřinec Hill, near Domažlice; Poběžovice; Schlaggenwald; Havírky near Pisek. *Moravia:* Viden; Vienna, Gross-Meseritsch; Cyrillhof, Gross-Meseritsch; Punau, Marienbad; Königswart, Marienbad (2 localities). *Silesia:* Peilau, Reichenberg.
- Norway:* Brevig; Landaas, Iveland. *Sweden:* Horrsjöberg, Wernmland; West of Lake Skruppetorp, Östergötland; Lilla Elysjö, Krockek, Linköping; Varuträsk. *Finland:* Sukula, Tammela; Viitaniemi, Eräjärvi parish, near Oriväsi; Lemnäs, Kimito; Mattkärr, Kimito; Helsingfors.
- Southwest Africa:* Karibib District; Erongo Area; Sandamab; Otjimboyo; North of Arandis. *Southern Rhodesia:* Tchetchenini Hill, Lomagundi. *Cape Province:* Wolf Kop, northeast of Kenhardt.
- Siberia:* Djidinskoe, Western Baikal. *India:* Banekhap, Singar, and Abraki Pahar, Pichhli, all in the Gaya District, Bihar Province, Bengal. *Korea:* Denso-men District, Heian-hokudo.

PARAGENESIS

Triplite occurs chiefly in pegmatites but also to a limited extent in hydrothermal veins of the high temperature variety. The occurrences may be grouped into the following types:

I. Pegmatites.

- A. Granitic pegmatites with a relatively simple and subordinate hydrothermal assemblage.
- B. Granitic pegmatites with a well developed phosphate assemblage.
- C. Granitic pegmatites characterized by a lithium phase.
- D. Granitic pegmatites with a tin-lithium phase.

II. High temperature veins.

A. Tin veins.

B. Tungsten veins.

C. Phosphatic veins with Al and Ti minerals.

Granitic pegmatites in which the hydrothermal mineral assemblage is relatively simple may contain triplite. Examples are Córdoba, Argentina, Skrupetorp, Sweden, and Eight Mile Park, Colorado. Mineral associates of the triplite are albite, beryl, muscovite, sulfides, tourmaline, and columbite.

Occurrences of triplite in the more complex phosphatic type of pegmatite are represented by the Bavarian and French localities, Mangualde in Portugal, Domažlice and Poběžovice in Bohemia, the Gaya district of Bengal, and Stoneham, Maine. Triphylite, apatite, and many other phosphates may accompany triplite. Columbite, beryl, sulfides, muscovite, and tourmaline are other typical associates. Minor cassiterite may be present, but both Sn and Li are subordinate.

Triplite-bearing lithium pegmatites with lepidolite and either or both spodumene and amblygonite occur in the Black Hills, near Wickenburg in Arizona, at Haddam Neck, Connecticut, and at Tammela, Finland. Various other phosphates usually are present as are beryl, tourmaline, muscovite, and tantalum minerals.

In the Karibib and Erongo areas of Southwest Africa occur triplite-bearing pegmatites characterized by cassiterite, lepidolite, tourmaline, wolframite, tantalite, and muscovite. The pegmatite assemblage at Lemnäs, Finland is similar, and the Geyer, Saxony occurrence also belongs here.

Only six vein occurrences of triplite have been recorded. The Korean deposit at Heian-hokudo (Ishibashi, 1944) is a tin-tungsten deposit with cassiterite, teallite and stannite, wolframite and scheelite, and the bismuth minerals, bismuthinite, native bismuth, and cosalite. Molybdenite and several other common sulfides also are present. Other tin veins that contain triplite occur in Bolivia (Ahlfeld, 1926) and at Schlaggenwald, Bohemia. At the former cassiterite, stannite, molybdenite and other sulfides appear; at the latter cassiterite and molybdenite also are found. Triplite occurs in tungsten veins near Aurum, Nevada and at Camp Signal, near Goffs, California. Huebnerite is the tungsten mineral common to both deposits. The Nevada occurrence also has scheelite and the bismuthinite-native bismuth-cosalite assemblage.

The Horrsjöberg, Sweden, occurrence of triplite ("talc-triplite") is unique. Quartz veins contain svanbergite, lazulite, apatite, triplite, pyrophyllite, damourite, rutile, ilmenite, garnet, tourmaline, and pyrite (Igelstrom, 1883).

In pegmatites triplite is usually the product of hydrothermal solutions and forms by replacement, for commonly it is associated with sodic plagioclase in secondary structural units. Apparently, it is developed during the early stages of the hydrothermal phase. However, Pehrman (1945) has placed it in the magmatic stage. The mineral has not been observed in vugs, but large, rough, partly faced crystals are recorded from three localities:

- (1) Hagendorf, Bavaria (Laubmann and Steinmetz, 1920).
- (2) Lemnäs, Kimito, Finland (Pehrman, 1939).
- (3) Mica Lode, Eight Mile Park, Fremont Co., Colorado (Wolfe and Heinrich, 1947).

Within a single pegmatite district triplite may show a very marked variation in composition, as for example, in Colorado (Anal. 1-3); Arizona (Anal. 8 and 9); and Kimito, Finland (Anal. 33 and 34). In some other districts the composition is relatively uniform: Bavaria (Anal. 19-21 and Anal. 16 and 17); Königswart, Marienbad, Moravia (Anal. 25 and 26), and Argentina (Anal. 10-12).

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PRELIMINARY OBSERVATIONS ON THE LUMINESCENCE ACTIVATION OF ZEOLITE MINERALS BY BASE EXCHANGE*

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ABSTRACT

Luminescence may be artificially induced in natural zeolite minerals by introducing activator elements (Mn, Pb, Ag, Cu) through base exchange. Base-exchanged Cu-zeolites and Ag-zeolites show green and blue cathodoluminescence, respectively. After dehydration, some Cu- and Ag-zeolites also develop photoluminescence, under ultraviolet excitation. Mn-, Pb-, and (Mn+Pb)-zeolites, which in their normal hydrated state are unresponsive to any excitation source, also develop cathodoluminescence and/or photoluminescence after being dehydrated.

Development of luminescence in base-exchanged zeolites is a reversible process dependent on the state of hydration or dehydration. The open-ness of the zeolites' crystal framework and its structural water are significant.

INTRODUCTION

It seems somewhat surprising, in view of their remarkable base exchange properties, that natural zeolite minerals do not luminesce; the literature records only one instance (Engelhardt, 1912), a specimen of natrolite from Oberscheffhauser, Germany, which fluoresces yellowish white under 3650Å excitation. The authors have found that luminescence may be artificially induced in natural zeolite minerals by the introduction of activator elements through base exchange.

The zeolites form an unusual group of hydrous aluminum silicates ($\text{Al}:\text{Ca}+\text{Na}=1:1$, $\text{Al}:\text{Si}:\text{O}=1:2$), which according to Dana (1932) are closely related in composition and occurrence. When heated, water is readily and continuously given off. This is in marked contrast to the normal release of water from minerals in discontinuous steps of definite amounts of water at definite temperature levels. The partially dehydrated zeolite can take up again its lost share of water, or even replace it by other substances such as air, ammonia, alcohol, hydrogen sulfide, iodine or mercury vapor. Moreover, the alkali or alkaline earth metal (usually Na and/or Ca) present may be replaced artificially by other metals, such as Ag, K, etc., by soaking in aqueous solution.

The atomic structure of the zeolite crystals (Wells 1945, Barrer 1949) consists of a more or less open framework of SiO_4 tetrahedra (with Al

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substituting for as much as half of the Si atoms), and linked together by shared oxygen atoms. The excess negative charge resulting from substitution of Al^{+3} for Si^{+4} allows for the introduction of positive ions (Ca^{++} or Na^{+}) into the cavities. Due to lattice defects in real crystals, these cations may move throughout the whole volume of the crystal rather than remain in a fixed position. In analcite (Taylor 1930), for example, 16 Na^{+} ions may occupy any of 24 vacancies. It is probable that the cations show six-fold coordination in all the zeolites.

The molecules of water (Taylor 1930, Wells 1945, Eitel 1941) are accommodated in the wide channels of the lattice, the number of molecules present being limited by the specific structure. Because of the electrostatic charge distribution in the water molecules they are found to require definite sets of nearest neighbors. In natrolite (Taylor, Meek, and Jackson 1932), the water molecules and oxygen atoms of the tetrahedron framework form double columns parallel to the c axis and passing through each channel; each of the 16 Na^{+} ions in the unit cell being surrounded by four oxygen atoms and two water molecules. Zeolitic water, then, is truly "structural" water.

Three types of zeolite structures can be distinguished: the analcite type, which contains a robust tri-dimensional lattice of tetrahedra cross-linked into 4- and 6-membered rings; the heulandite type, which is lamellar, consisting of close-knit sheets or layers of tetrahedra; and the natrolite type, which has chains of tetrahedra with only few cross-linkages, giving rise to the fibrous zeolites.

EXPERIMENTAL TECHNIQUE

Preliminary studies have been made with the following zeolites, which were kindly supplied us by Dr. William Foshag of the U. S. National Museum: chabazite (Pennsylvania), heulandite (Essex Co., N. J.), natrolite (Moore's Station, N. J.), and stilbite (Essex Co., N. J.). All three lattice structure-types of zeolites are represented, chabazite having the tri-dimensional network, heulandite and stilbite having the layer lattice, and natrolite, the fibrous structure. Most of our detailed work was based on stilbite since this was available in larger quantities.

Before treatment, all the mineral specimens were inspected under ultraviolet and cathode ray excitation¹ and found to be non-luminescent.

¹ The "Mineralite," a low pressure mercury-vapor lamp of the cold cathode type, fitted with a Corning #9863 filter (to absorb visible blue light) emits primarily the mercury 2537Å resonance line. Sylvania's "Blacklite" bulb with a #9863 filter was used as a source of 3650Å radiation. Electron bombardment was supplied by a special demountable cathode ray tube or by spark coil discharge in partial vacuum.

A sample of each mineral was then examined spectrographically for conspicuous impurity elements. These are listed in the table following:

TABLE 1. MINOR AND TRACE IMPURITIES IN ZEOLITES

Chabazite:	Ti, V, Fe, Mn, Mg, Sr, Ba, Cu, ² Ag, ² (B)
Heulandite:	Ti, V, Fe, Mn, Mg, Sr, Ba, Cu, Ag, B
Natrolite:	Ti, V, (Fe), (Mn), Mg, Ca, (Cu), Ag, B
Stilbite:	Ti, V, (Fe), (Mn), Mg, (Sr), Cu, Ag, B

() indicates bare trace

The presence of trace impurities of Cu, Ag and Mn, so-called activator elements, does not invalidate the test results, since the zeolite minerals were completely unresponsive to all sources of excitation before treatment.

X-ray powder patterns were obtained for each zeolite mineral and compared with *d*-values (interplanar spacings) as listed in the American Society for Testing Materials' index of X-ray diffraction data. All gave reasonably good correlation.

Weighed samples of the powdered minerals (ground to pass 100 mesh sieve) were placed in glass-stoppered vials with a constant, measured volume of aqueous solution containing the desired concentration of activator. Unless otherwise specified, all preparations were made at room temperature. After treatment, the mineral powders were filtered off through sintered glass crucibles, washed with water and acetone, and air-dried.

LUMINESCENCE EFFECTS

In the earliest survey, samples of each of the zeolite minerals were treated for one week with 1×10^{-3} M aqueous solutions of the following metals (known to act as luminescence activators under suitable conditions): Cu, Ag, Mn, Pb, and Mn+Pb. None of the preparations luminesced under ultraviolet excitation, either 2537Å or 3650Å. Under electron bombardment from a spark coil discharge, however, the Cu-zeolites, with the exception of natrolite, luminesced a bright green. Later natrolite also was made luminescent after treatment with a more concentrated copper solution. The Ag-zeolites also responded to cathode ray excitation, with a blue-green luminescence (see Table 3).

Further studies were confined to Cu-activation. The effect of varied

² Copper impurity may have been introduced with the graphite powder which was added as a spectrographic "flux," or from the solder in the stainless steel-meshed sieve. Silver also may possibly be a foreign contaminant, from previous materials passed through the sieve.

concentrations of copper and varied immersion periods were investigated first. Stilbite was treated with copper solutions ranging in concentration from 1×10^{-6} M to 1 M, at ten-fold intervals, and with a saturated solution. For a study of the time factor, stilbite preparations were taken after 15 and 30 minutes; 1, 2, 4, 8, and 16 hours; 2 days and 8 days.

It was found that after as little as 15 minutes' immersion in 1×10^{-3} M Cu^{++} solution (maximum Cu available by base exchange = 15×10^{-4} g. per g. sample), stilbite developed its characteristic green luminescence under cathode ray excitation. On the basis of chemical evidence, Eichhorn (1859) likewise noted that considerable exchange with other ions took place within the first few minutes, although equilibrium was not established for some time. The lowest concentration to produce noticeable luminescence was 1×10^{-5} M Cu^{++} (maximum Cu available = 15×10^{-6} g. per g. sample), after treatment for one week. There were no appreciable differences in the luminescence intensity of the other preparations.³ A powdered sample of stilbite immersed in 1×10^{-3} M Cu^{++} solution in a sealed tube and heated at 100° C. for one hour did not develop noticeably brighter luminescence than the corresponding room temperature preparation.

In order to study more closely the nature of the luminescence emission of the Cu-zeolites, a bright-luminescing stilbite (treated with 1×10^{-3} M Cu^{++} , 1 week, room temperature) was selected for spectrophotometric measurement. Samples of the Cu-stilbite and the natural stilbite were set up within a special demountable cathode ray tube; the system was evacuated to a pressure of less than 2×10^{-5} mm. Hg and then subjected to electron bombardment. The luminescence emission spectrum was then determined photoelectrically using a motor-driven monochromator, a photomultiplier-tube, D. C. amplifier, and an Esterline-Angus Recorder. The emission spectrum is reproduced as Fig. 1.

DISCUSSION

Consideration of the available evidence leads us to believe that luminescence of the zeolites is connected with the entry of Cu^{++} ions into the crystal lattice structure and not with surface adsorption (as is the case with some organic phosphors in silica gel).

By spectrochemical analysis of a series of substituted stilbites, it was determined that approximately all of the Cu^{++} originally present in the less concentrated solutions was taken up by base exchange after one

³ Because of limitations inherent in the cathode ray equipment and the low luminescence intensity of the samples, it was not practicable to make exact measurements. Samples were inspected by eye, and only three or four at most could be compared simultaneously.

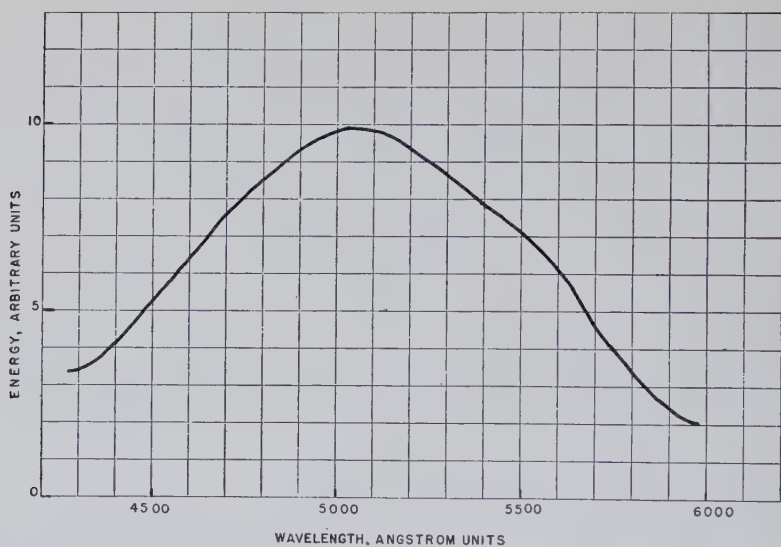


FIG. 1. Spectral emission of Cu-stilbite, under cathode ray excitation.

week at room temperature. Chemical analysis for residual Cu^{++} in the corresponding filtrates from these preparations confirmed the results.

TABLE 2. DETERMINATION OF CU IN SOME BASE EXCHANGED ZEOLITES

Sample Preparation	Cu added, as wt. % in zeol.	Cu found spectr., as wt. %	Cu found chem., as wt. %
Stilbite, nat'l.	None	$\leq .0001$ impurity	—
Stilbite in 10^{-4}M Cu^{++}	.002	.003	.002
Stilbite in 10^{-3}M Cu^{++}	.02	.05	.02
Stilbite in 10^{-2}M Cu^{++}	0.2	abt. 0.3	0.13
Stilbite in 10^{-1}M Cu^{++}	2.0	abt. 1.	1.

Zoch (1914) conducted base exchange studies with copper, but its valence state was not specifically indicated. Judging from the fact that her Cu-substituted zeolites were colored green, it is assumed to have been the cupric ion. Those of our zeolite samples which had been treated with the more concentrated Cu^{++} solutions, 1×10^{-2} M and higher, also became green in color. Zoch found, furthermore, that the green coloration could be discharged by soaking the specimen in ammonium chloride solution, the Cu^{++} ion being reexchanged for NH_4^+ as shown by chemical analysis. There was a definite quantitative relationship between the amounts of replaced Na^+ , Cu^{++} , and NH_4^+ .

Similarly, we were able to discharge the green color of a Cu-stilbite sample by digesting with hot sodium chloride solution. The colored Cu-stilbite luminesced bright green; the decolorized material (Na-stilbite) was dead. Chemical tests established that Cu^{++} had been removed from our green-colored zeolite and had been transferred to the leaching solution.

To eliminate the possibility that the observed luminescence phenomena were surface adsorption effects, experiments were carried out with silica gel, amorphous silica (H_2SiO_3), and finely powdered alumina. Treatment with Cu^{++} solutions produced completely negative results.

X-ray powder patterns showed no change in the lattice dimensions of the Cu-zeolites. That lattice distortion does occur in substituted zeolites has been demonstrated by the researches of Taylor, Meek and Jackson (1932) and Hey and Bannister (1932), among others. Their studies were made with *completely* base-exchanged zeolites. Since our prime interest was luminescence activation, no special effort was undertaken to attain complete base exchange. It is probable that immersion at room temperature in the very dilute solutions used effected only partial exchange. Where concentrated solutions were employed it is likely that partial decomposition of the zeolite (by the free acid resulting from unavoidable hydrolysis of the copper salt) deposited a protective coating of silica around the mineral grains, which then prevented extensive base exchange.

It may be of interest at this point to call attention to Hey and Bannister's (1932) statement that natrolite does not take up Cu^{++} by base exchange even after four months' immersion at room temperature. Their criteria were optical and X-ray behavior. That luminescence activation is a much more sensitive test for base exchange is apparent from the fact that natrolite (treated with 10^{-2} M Cu^{++} , 1 week, room temperature) shows distinct green cathodoluminescence.

EFFECT OF DEHYDRATION AND REHYDRATION ON SUBSTITUTED ZEOLITES

The literature contains voluminous reports, some of it contradictory, on the peculiar dehydration properties of the zeolites, including some x-ray studies on the structural significance of water in the crystal lattice. Hey (1930), in reviewing the general properties of the zeolites states: "In some zeolites the water may probably be all removed without any discontinuity at all; as dehydration proceeds, there is a tendency for the whole structure to shrink somewhat, provided the temperature is maintained high enough for a long enough period, and after such shrinkage, the reabsorption of water is prevented, or at least slowed. If, however,

TABLE 3. LUMINESCENCE RESPONSE OF BASE-EXCHANGED ZEOLITES

Sample	Hydrous Preparations				Dehydrated (Heated) Preparations												Cathode Ray
	3650 Å		2537 Å		3650 Å				2537 Å				2537 Å				
	3650 Å	2537 Å	Cathode Ray		Fresh	Aged	Refired	Rehydr.	Fresh	Aged	Refired	Rehydr.	Fresh	Aged	Refired	Rehydr.	
Chabazite, nat'l Heulandite, nat'l Natrolite, nat'l Stilbite, nat'l	dead	dead	dead	dead	gray	dead	dead		gray	dead			gray	dead			dead
	dead	dead	dead	dead	whitish	dead	dead		whitish	dead			whitish	dead			dead
	dead	dead	dead	dead	whitish	dead	dead		whitish	dead			whitish	dead			dead
	dead	dead	dead	dead	whitish	dead	dead		whitish	dead			whitish	dead			dead
Chabazite: Mn Heulandite: Mn Natrolite: Mn Stilbite: Mn	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	v. pale pink
	dead	dead	dead	dead	dead (?)	dead	dead	dead	dead (?)	dead	dead	dead	dead	dead	dead	dead	pale pink
	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	white
	dead	dead	dead	dead	pink	dead	dead	dead	pink	dead	dead	dead	dead	pink	dead	dead	v. pale pink
Chabazite: Pb Heulandite: Pb Natrolite: Pb Stilbite: Pb	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead
	dead	dead	dead	dead	whitish	dead	dead	dead	whitish	dead	dead	dead	whitish	dead	dead	gray	blue
	dead	dead	dead	dead	bluish-white	dead	dead	dead	bluish-white	dead	dead	dead	whitish	dead	gray	gray	bluish-white
	dead	dead	dead	dead	bluish-white	dead	dead	dead	bluish-white	dead	dead	dead	whitish	dead	whitish	dead	dead
Chabazite: Mn + Pb Heulandite: Mn + Pb Natrolite: Mn + Pb Stilbite: Mn + Pb	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dull pink	dull pink	dull pink	pink
	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dull pink	dull pink	pale pink
	dead	dead	dead	dead	bluish-white	dead	bluish-white	gray	pale blue	dead (?)	whitish	gray	whitish	pale pink	gray	gray	bluish-white
	dead	dead	dead	dead	bluish-white	dead	bluish-white	gray	pink	pale pink	pink	pink	whitish	pale pink	dull pink	dull pink	pink
Chabazite: Ag Heulandite: Ag Natrolite: Ag Stilbite: Ag	dead	dead	dead	dead	dead	dead	dead	dead	white	dead	blackened on standing		white	dead	blackened on standing		green
	dead	dead	dead	dead	white	dead	blackened on standing		white	dead	blackened on standing		white	dead	blackened on standing		dead (?)
	dead	dead	dead	dead	white	dead	blackened on standing		white	dead	blackened on standing		white	dead	blackened on standing		blue
	dead	dead	dead	dead	white	dead	blackened on standing		white	dead	blackened on standing		white	dead	blackened on standing		dead
Chabazite: Cu Heulandite: Cu Natrolite: Cu Stilbite: Cu	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	dead	green
	dead	dead	dead	dead	dead	dead	dead	dead	pale green	dead (?)	pale green	dull green	pale green	dead	dull green	dull green	green
	dead	dead	dead	dead	dead	dead	dead	dead	white	dull white	dead (?)	white	white	dead	white	bluish-white	green
	dead	dead	dead	dead	dead	dead	dead	dead	white	dull white	white	white	white	dead	white	bluish-white	green

the dehydration is carried out at low temperature in vacuo, the lattice-shrinkage ('Gitterreaktion') of Rinne and Scheumann is largely or entirely avoided, and the rehydration is rapid." The completely dehydrated zeolites, or "metazeolites" (high-temperature forms), still retain structural resemblance to the parent material, although x-ray patterns may indicate shrinkage or "collapse" of the unit cell.

It is impossible to effect extensive base exchange in a completely dehydrated zeolite. Beutell and Blaschke (1915) believe that removal of the water decreases the mobility of the cations. This loss of mobility may be due, they suggest, to the lattice shrinkage and/or increased electrical attraction between ions after the removal of the highly polar water molecules.

In an attempt to study the effect, if any, of the structural modifications produced by dehydration on the luminescent properties of base-exchanged zeolites, the cathodoluminescent Ag- and Cu-zeolites, as well as the non-luminescent Mn-, Pb-, and (Mn+Pb)-substituted zeolites, were subjected to thermal treatment. After heating for 30 minutes at 300° C. in a muffle-type furnace and cooling to room temperature, several of these base-exchanged zeolites, including some of the previously inert samples, luminesced under 2537Å and/or 3650Å excitation. Natural zeolite minerals which had not undergone base exchange but only thermal treatment were found to show indications of feeble photoluminescence.

Upon re-examination some eight months later, it was noted that the luminescence had weakened considerably and even disappeared completely from some of the heat-treated zeolites. After another heat treatment, however, luminescence was restored to these samples, although not all were returned to their original intensity. The entire suite of samples was subsequently stored over water in desiccators. After two weeks, the luminescence had again noticeably dimmed. This reversible development and loss of luminescence is obviously related to the hydrous condition of the zeolite mineral structure.

Table 3 lists the luminescence response of the various zeolite preparations studied.

Our most recent experiments indicate interesting possibilities in developing zeolites which will both fluoresce and phosphoresce under ultraviolet excitation with intensity comparable to efficient commercial phosphors. This has been accomplished by certain variations in thermal treatment and will be reported at a later date.

SUMMARY

The luminescence data can be correlated with the hydrous condition of the zeolites and summarized, as follows:

(1) Natural (normally hydrated) zeolite minerals do not luminesce under any excitation source but after heating at 300° C (dehydrated, to an undetermined extent), the zeolites develop indications of weak white luminescence under 2537Å and 3650Å excitation. (Some impurity in the mineral may be responsible for the weak activation.)

(2) Base-exchanged zeolites in their fully hydrated state (our room temperature preparations) do not exhibit photoluminescence, and only the Cu- and Ag-zeolites show cathodoluminescence.

(3) Some of the same base-exchanged zeolites dehydrated (to an undetermined degree) by heating at 300° C develop photoluminescence and/or cathodoluminescence. Specifically, the Mn-zeolites become responsive to cathode ray excitation; some of the Pb-zeolites respond to all excitation sources; of the (Mn+Pb)-zeolites, all respond to cathode ray and some to ultraviolet excitation as well; and some of the Cu- and Ag-zeolites become responsive to ultraviolet excitation.

(4) When these same heated, base-exchanged, luminescent zeolites are rehydrated (either by aging in the open or storing in an atmosphere of water vapor) they lose their luminescence activity, wholly or partially.

(5) The development of luminescence in base-exchanged zeolites is a reversible reaction and can be produced or suppressed directly by a comparable shift in the dehydration-rehydration process (within the limits noted in the literature).

(6) Since the zeolite minerals contain different amounts of structural water, show different dehydration rates and reach complete dehydration at different temperatures, it is to be expected that their correlated luminescence development should likewise differ in rate and extent.

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LOW TEMPERATURE LIQUID IMMISCIBILITY IN THE SYSTEM $K_2O-FeO-Al_2O_3-SiO_2$

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ABSTRACT

Liquid immiscibility has been found in anhydrous silicate melts at temperatures as low as 1100° C and in compositions containing as much as 16% alumina plus alkalis in the system $K_2O-FeO-Al_2O_3-SiO_2$. A preliminary equilibrium diagram is presented for the system leucite-fayalite-silica, a plane through the more general system, which cuts the new region of immiscibility.

Some students of petrology have considered the occurrence of certain rock pairs in the field to indicate splitting of rock magmas due to liquid immiscibility. Such an association, however, may be explained with equal facility by other processes. In 1927 Greig* published the results of his experimental equilibrium studies on a number of silicate systems showing that liquid immiscibility does indeed occur in some instances, but that the high temperatures and limited compositions involved were such that the occurrence of the process in nature was unlikely. During the course of phase equilibrium studies on the quaternary system $K_2O-FeO-Al_2O_3-SiO_2$, the writer has found an entirely unexpected area of liquid immiscibility (completely surrounded by miscibility), at much lower temperatures and less restricted compositions. This discovery lightens somewhat the burden of proof for the advocate of magma splitting, but does not affect the main objection which Bowen (1928) raises, that the mechanism of liquid immiscibility is such that evidence of its occurrence in nature should be visible in the glassy rocks; this evidence has not been found.

Work is still in progress on the system, but as the discovery of this new area is of considerable interest, and as it will be some time before sufficient data can be obtained to present a completed equilibrium diagram for this part of the system, a preliminary diagram will be given here, based on available data. Figure 1 represents the ternary subsystem leucite ($K_2O \cdot Al_2O_3 \cdot 4SiO_2$)-fayalite ($2FeO \cdot SiO_2$)- SiO_2 . It can be considered as a plane through the tetrahedron representing the more general quaternary system $K_2O-FeO-Al_2O_3-SiO_2$. The data were obtained by the method of quenching, using essentially the techniques of Bowen and Schairer (1932). All melts were run in intimate contact with pure metallic iron, thus containing only that amount of Fe_2O_3 which represents equilibrium with metallic iron at the liquidus temperature for the individual melt composition. Each melt was analyzed for ferrous and ferric iron after such treatment, and the ferric iron content recalculated to ferrous in

* References are given at the end of the paper.

order to plot the diagram. The variation of the equilibrium constant $[Fe_2O_3]/[FeO]$ with temperature and composition of the liquid phase is such as to yield from 0.5 to 2.5 weight per cent Fe_2O_3 in most of the compositions studied. The diagram is based on approximately 500 quenching runs made on 26 compositions; considerably more data are

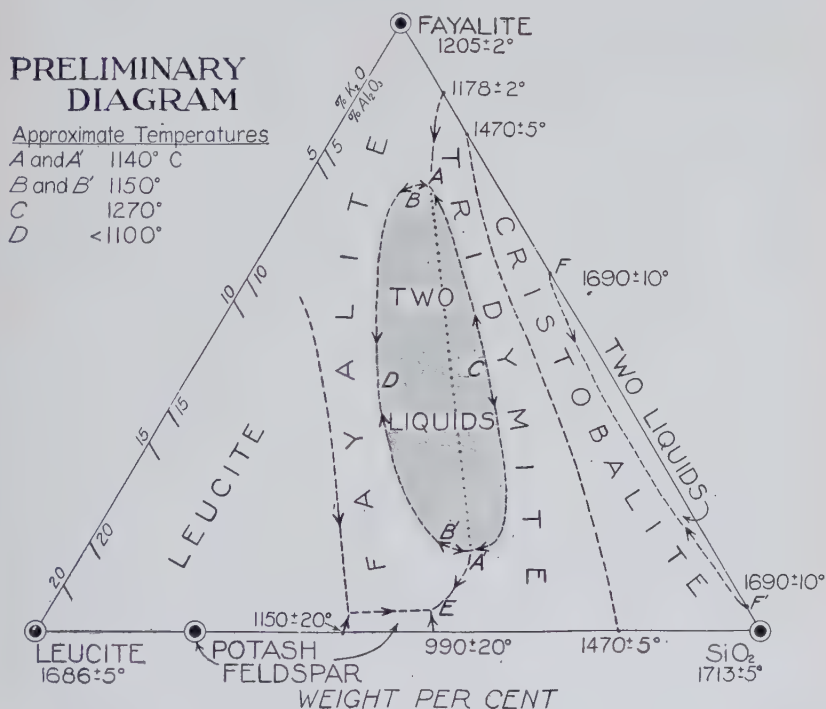


FIG. 1. Preliminary equilibrium diagram for the system leucite-fayalite-silica, showing the fields of the several crystalline phases and the intersection of the new area of immiscibility with the liquidus surface (shaded). In the area $AA'CA$ two liquids are in equilibrium with silica crystals, and in the area $AA'B'DBA$ two liquids are in equilibrium with fayalite crystals. Along the line AA' two liquids (A and A') are in equilibrium with each other and with crystals of both silica and fayalite. All melts contain Fe_2O_3 in amounts representing equilibrium with pure metallic iron.

needed, and when these are obtained a more detailed paper will be presented, giving composition and quench data as well as experimental technique. The data on the sidelines fayalite-silica and leucite-silica (Fig. 1) are from the published works of Bowen and Schairer (1932) and Schairer and Bowen (1947).

The new area of liquid immiscibility has been indicated in Fig. 1 by shading (the area $ACA'DA$). Greig (1927) found liquid immiscibility at

high temperatures in the system FeO-SiO_2 (of which the binary system fayalite-silica is a portion), and there must be an extension of this region of liquid immiscibility into the quaternary system $\text{K}_2\text{O-FeO-Al}_2\text{O}_3\text{-SiO}_2$. In the plane studied, this extension is evidently small, as indicated by the dashed line between F and F' , and the newly discovered area of two liquid formation appears to be entirely separate from it. The new area may be divided into two portions. In the area $AA'CA$ two liquids are in equilibrium with tridymite crystals (in part metastably with cristobalite in place of tridymite), and in the area $AA'B'DBA$ two liquids are in equilibrium with fayalite crystals. Liquids A and A' are in equilibrium with each other and with both tridymite and fayalite crystals, and hence represent invariant conditions (neglecting the effect of the Fe_2O_3 as another component). AA' is the only line connecting conjugate liquids that can be drawn at present with any degree of certainty, although it appears that the other conjugation lines will be roughly parallel to it. Present data would seem to indicate that there are also a pair of maximum points B and B' , somewhere along the line ADA' , joining conjugate liquids in equilibrium with fayalite crystals, with temperatures dropping away toward D and AA' on both sides of these maximum points; the true positions of these two maximum points B and B' on the line ADA' are not known. The true positions of point C , representing the maximum temperature for the existence of two liquids in equilibrium with crystals of silica, and point D , representing the minimum temperature for the existence of two liquids in equilibrium with crystals of fayalite (and the minimum temperature for the existence of two liquids), are also unknown at present.

Liquid A , when quenched to a glass, is dark greenish brown, with an index of refraction of approximately 1.70, and liquid A' is colorless, with an index of 1.51. Microscopic observation of such samples, showing two liquids (two glasses), each embedded in and containing globules of the other, is striking but is not the only evidence for liquid immiscibility. More significant perhaps is the thermal evidence of two liquids of grossly different composition (A and A') both in equilibrium with fayalite and tridymite crystals, at the *same* temperature, within the limits of variation imposed by the presence of the small, equilibrium amounts of ferric iron and the experimental error in measurement. High viscosities and difficulties inherent in the experimental technique necessary for silicate equilibrium studies involving ferrous oxide have precluded obtaining samples of individual liquids for analysis, but experiments with that objective are in progress. None of the points or lines on Fig. 1 are sufficiently well established in composition to be located definitely, but the shape of the

diagram is not expected to be changed radically by further data. Present data establish the position of ACA' with far better definition than ADA' .

This area of immiscibility as shown on Fig. 1 actually represents only the intersection of a liquid immiscibility "dome" with the liquidus surface, i.e., two liquids in equilibrium with crystals of fayalite or silica or both. At higher temperatures, batches having gross compositions falling in the area $ACA'DA$ will consist of two liquids alone. Insufficient data are available to draw isotherms on the surface bounding this region of two liquids, but they are adequate to show that immiscibility is eliminated by a comparatively small increase in temperature in the compositions studied, none of them exhibiting two liquid formation above approximately 1260°C .

Compositions showing immiscibility are not confined to this plane section through the tetrahedron representing the more general system $K_2O-FeO-Al_2O_3-SiO_2$, as adjacent compositions have been made and they also show it. The plane of Fig. 1 thus represents only a section through an immiscibility volume. The system $FeO-Al_2O_3-SiO_2$ (one side of the tetrahedron) shows no such region of low temperature immiscibility, thus indicating that the presence of K_2O is necessary in this case, but the effect of the addition of other materials such as Na_2O or H_2O on the extent of immiscibility is unknown. For this reason it is impossible at present to apply these results directly to the problem of the origin of those special rock types that approach most closely the compositions studied. Liquid A' can be considered to be equivalent to a fayalite bearing potash-granite, although the eutectic point E (the exact composition of which has not been determined) would represent a closer approach to the composition of some natural fayalite granites, neglecting the fact that these tend to be soda-rich rather than potash-rich. This does not require, however, that these rocks originated through the process of liquid immiscibility. More significant perhaps for the proponent of immiscibility is the fact that it can occur at temperatures as low as 1100°C in dry silicate systems containing as much as 16% alumina plus alkali,* but the problem of satisfactory evidence for it in the rocks looms as large as ever.

* Point B' (Fig. 1) as drawn could be represented in the following terms:

K_2O	7.8%	Leucite	36%	Potash Feldspar	46%
Al_2O_3	8.4%	SiO_2	50%	SiO_2	40%
FeO	9.9%	Fayalite	14%	Fayalite	14%
SiO_2	73.9%				

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ALIGNMENT CHART FOR CALCULATION OF REFRACTIVE INDEX FROM THE DEVIATION OF LIGHT BY A PRISM

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ABSTRACT

An alignment chart is presented by means of which the refractive index of a prism is obtained from the angle of deviation of light passing through it, and the prism angle.

New interest in the prism methods for measurement of refractive indices has resulted from the development of new designs for goniometers (Wolfe and Franklin, 1949; Wolfe, 1948). Such interest suggests that the prism methods and computations therefor might profitably receive additional study. This note describes an improved nomogram for the computation. Smith (1906) described a chart for use with the minimum deviation method, accurate to the nearest .005. Other reductions of equations (1) or (2) to chart form are not known to the writer.

Equations (1) and (2) are used for computing the refractive index, n , from measurements of the prism angle, a , and the deviation, d , of a light beam passing through it.

$$n = \frac{\sin (a + d)}{\sin a} \quad (1)$$

$$n = \frac{\sin \frac{(a + d)}{2}}{\sin \frac{a}{2}} \quad (2)$$

It is obvious that any nomogram for one will also give solutions of the other, provided only that the scales for a and d have two sets of numerical calibrations differing by a factor of two. Equation (1) is the form used when the light enters (or leaves) one side of the prism at normal incidence (Ford, 1932, p. 240); equation (2) is the form used when the deviation of light is minimum (Ford, 1932, p. 242).

These equations may be solved by means of the alignment chart reproduced in Fig. 1. A straight line passing through the scales a and d at points corresponding respectively to the prism angle and to the angle of deviation will pass through the n -scale at a point corresponding to the refractive index of the prism. It is to be noted that the inner calibrations of the a -, and d -scales apply to the case of normal incidence of the light rays upon one prism face, and the outer (*slant-lettered*) calibrations, to the case of minimum deviation.

The coordinates of the various points along each scale have been obtained for approximately twice as many points as are reproduced here,

and when plotted on a chart measuring 100 cm. by 80 cm., readings of refractive index can easily be obtained that are accurate to the nearest 0.003, or better, from angular data accurate to the nearest minute of arc.

DERIVATION OF THE ALIGNMENT CHART

Equation (1) can be rewritten as follows:

$$n \sin a - \sin a \cos d - \cos a \sin d = 0 \quad (3)$$

In determinant form, this is:

$$\begin{vmatrix} n & 0 & 1 \\ \cos a & \sin a & 0 \\ \cos d & -\sin d & 1 \end{vmatrix} = 0$$

Rearrangement of the determinant gives the following standard form (Mackey, 1944, p. 85):

$$\begin{vmatrix} 0 & \tan a & 1 \\ 1/n & 0 & 1 \\ \sec d & -\tan d & 1 \end{vmatrix} = 0 \quad (4)$$

Equation (4) is recognized as the equation of a straight line through the three points, $x_1=0, y_1=\tan a$; $x_2=1/n, y_2=0$; and $x_3=\sec d, y_3=-\tan d$. It will be noted that the first point is determined solely by the prism angle a , the second by the refractive index n , and the third by the angle of deviation d . The first lies along the positive y -axis, the second, along the positive x -axis; the third, along a curve in the quadrant of positive x , negative y (Fig. 2A).

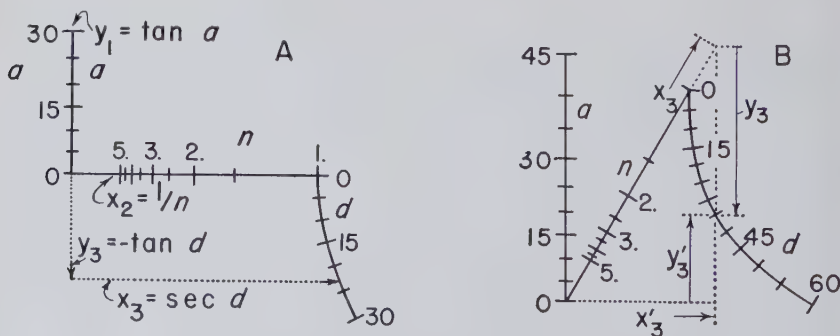


FIG. 2

The angle between the x - and the y -axis does not have to be 90° . The value 30° would give a satisfactory chart which is sketched in Fig. 2B. To facilitate plotting the d -scale on ordinary graph paper, the rectangular coordinates (x'_3, y'_3) of the scale points may be found after the above choice of 30° for the angle between the x - and the y -axis is settled (equation

CHART FOR DETERMINING REFRACTIVE INDEX From Deviation of Light by a Prism

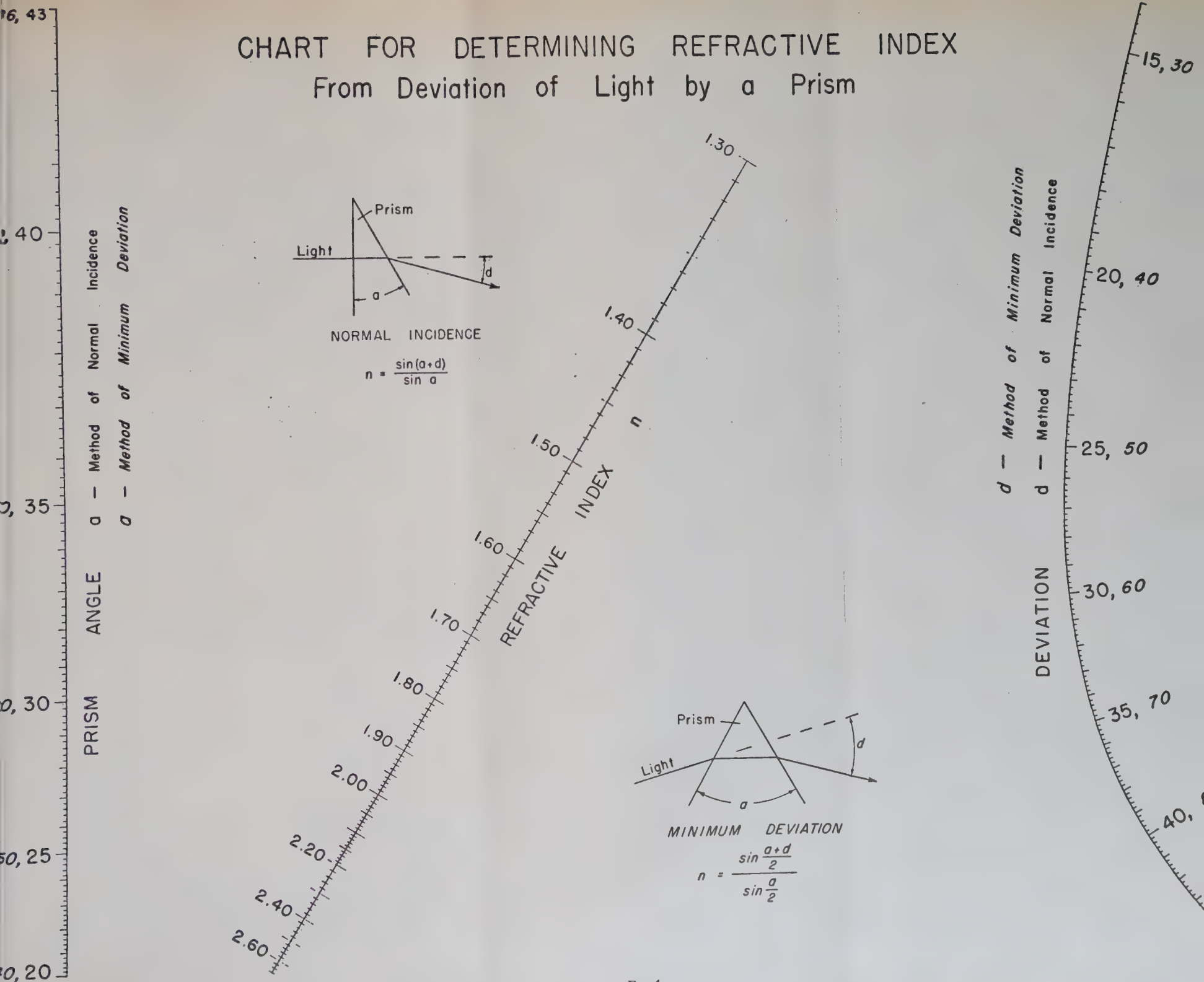


FIG. 1

5), or the original coordinates (x_3, y_3) , measured *parallel to the axes at the new angle of 30°* may be used as suggested in the figure.

$$\left. \begin{aligned} x_3' &= \sin 30^\circ \sec d \\ y_3' &= \cos 30^\circ \sec d - \tan d \end{aligned} \right\} \quad (5)$$

No computing whatever is necessary for the scales of a and n ; the x_3 and y_3 values for the scale of d may likewise be found in any table of tangents and secants, but the more convenient rectangular coordinates x_3' and y_3' must be computed.

A projective transformation of equation (4) (Mackey, 1944, p. 97) produces a more useful chart, and at the same time gives it a better shape with selected points of the a -, and d -scales at the corners of a square or rectangle. This transformation is accomplished by multiplying both sides of equation (4) by a determinant (6) in which the a 's, b 's, and c 's are constants to be evaluated.

$$\begin{vmatrix} a_1 & b_1 & c_1 \\ a_2 & b_2 & c_2 \\ a_3 & b_3 & c_3 \end{vmatrix} \neq 0 \quad (6)$$

The product is given as follows:

$$\begin{vmatrix} \frac{b_1 \tan a + c_1}{b_3 \tan a + c_3} & \frac{b_2 \tan a + c_2}{b_3 \tan a + c_3} & 1 \\ \frac{a_1 + nc_1}{a_3 + nc_3} & \frac{a_2 + nc_2}{a_3 + nc_3} & 1 \\ \frac{a_1 \sec d - b_1 \tan d + c_1}{a_3 \sec d - b_3 \tan d + c_3} & \frac{a_2 \sec d - b_2 \tan d + c_2}{a_3 \sec d - b_3 \tan d + c_3} & 1 \end{vmatrix} = 0 \quad (7)$$

We may choose to make the alignment chart 1 meter square, with the a -scale along the left edge ranging from 20° to 45° , and the d -scale curving inward from the right edge and ranging from 10° to 45° . Since calculations are simplified by taking the range of the a -scale to 45° , this square shape was selected, though the upper fifth of the square is omitted in Fig. 1. Placing the origin of coordinates at the lower left corner, we obtain the following eight equations:

Origin: $(x_1=0, y_1=0)$

$$x_1 = \frac{b_1 \tan 20 + c_1}{b_3 \tan 20 + c_3} = 0 \quad y_1 = \frac{b_2 \tan 20 + c_2}{b_3 \tan 20 + c_3} = 0$$

Upper left corner $(x_1=0, y_1=1.00 \text{ meter})$:

$$x_1 = \frac{b_1 \tan 45 + c_1}{b_3 \tan 45 + c_3} = 0 \quad y_1 = \frac{b_2 \tan 45 + c_2}{b_3 \tan 45 + c_3} = 1$$

Upper right corner $x_3=1, y_3=1$

$$x_3 = \frac{a_1 \sec 10 - b_1 \tan 10 + c_1}{a_3 \sec 10 - b_3 \tan 10 + c_3} = 1 \quad y_3 = \frac{a_2 \sec 10 - b_2 \tan 10 + c_2}{a_3 \sec 10 - b_3 \tan 10 + c_3} = 1$$

Lower right corner $(x_3=1, y_3=0)$

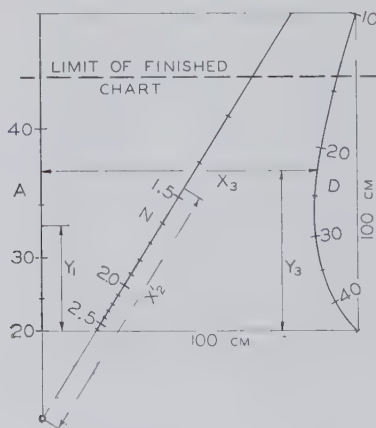
$$x_3 = \frac{a_1 \sec 45 - b_1 \tan 45 + c_1}{a_3 \sec 45 - b_3 \tan 45 + c_3} = 1 \quad y_3 = \frac{a_2 \sec 45 - b_2 \tan 45 + c_2}{a_3 \sec 45 - b_3 \tan 45 + c_3} = 0$$

These eight equations contain nine unknown quantities of which one

(c_3) is arbitrarily put equal to unity, reducing the system to eight equations in eight unknowns. Inspection shows that $b_1 = c_1 = 0$, simplifying the system at once. The final solution gives the elements of the multiplying determinant (6) as follows:

$$\left. \begin{array}{lll} a_1 = 0.325911 & b_1 = 0 & c_1 = \\ a_2 = 0.726998 & b_2 = 0.753776 & c_2 = -0.274352 \\ a_3 = -0.749293 & b_3 = -0.520575 & c_3 = 1 \end{array} \right\} \quad (9)$$

These values are substituted into equation (7) for the construction of the final alignment chart, which is reproduced in Fig. 1. The two equations for the n -scale are known to lead to a straight scale, and can therefore be combined into a single equation of distance x_2' (Fig. 3) along the line from its point of intersection with the y -axis at $y = -0.27435$. As shown by the dashed line in Fig. 3, the upper fifth of the square is omitted. The finished chart is presented as Fig. 1.



PRESENTATION OF THE ROEBLING MEDAL OF THE
MINERALOGICAL SOCIETY OF AMERICA TO
NORMAN L. BOWEN

ARTHUR F. BUDDINGTON, *Princeton University, Princeton, New Jersey.*

Mr. President, Fellows and Members of the Mineralogical Society of America, and guests:

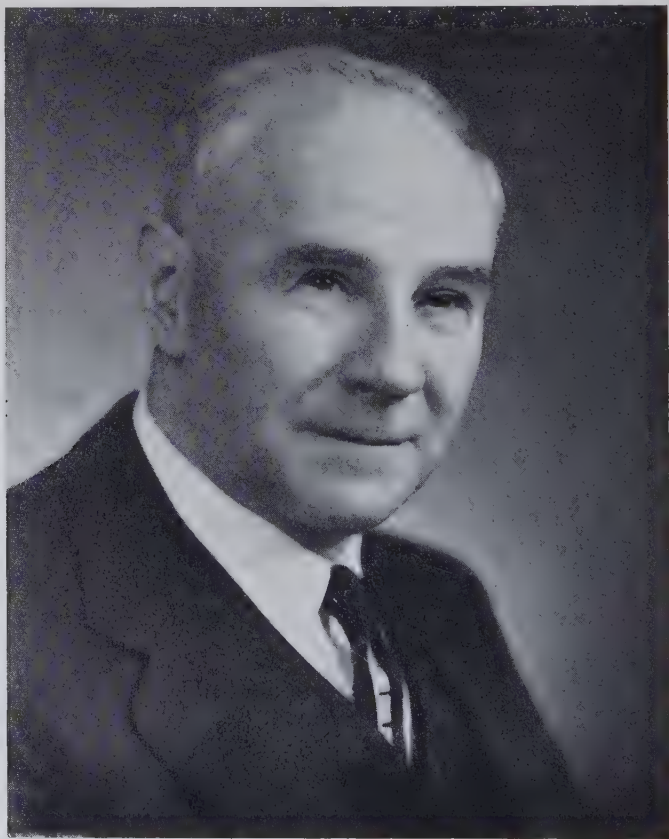
The Roebling medal is for meritorious achievement in the fields of the Mineralogical Sciences and is the highest honor awarded by the Mineralogical Society of America.

The award this year is the tenth since the initial one in 1937. For the first time it goes to a physicochemical petrologist and mineralogist, Dr. N. L. Bowen, whose researches have been based preeminently on *systematic quantitative* experimental physicochemical work, a field which has been exclusively developed in the present century. Dr. Bowen also further exemplifies a peculiarly significant 20th century development, a Canadian in the tradition of A. C. Lawson and R. A. Daly who has crossed to the United States and in addition to so outstandingly advancing the science has also helped to educate our students.

Before considering further the serious side of our medallist, I shall digress to speak of certain lighter matters. Professor Palache in telling something of the life and character of Colonel Roebling referred to his keenly developed sense of humor. Certainly, the Colonel would most highly approve of Dr. Bowen. May I quote some excerpts from his writings. The following is from an essay on "The Granite Problem and the Method of Multiple Prejudices"; "we hurl epithets, we hurl them back. We are most thankful that we are not as other men are. It may be that the mutual conflicts of prejudices will dull the rabid edge of each, and that the communal petrologic mind can eventually reach the truth by this sorry method," again "Pushed to its logical consequences for a large mass, that corollary of wet granitization, the 'basic front' is thus a basic affront to the intelligence of the geologic fraternity." In discussing the lack of significance of the eutectic relationships for magmatic differentiation he writes "whatever the conditions, the eutectic is the goal of all liquids; all attain it; none pass it; it is a fen of stagnant waters," and finally from his presidential address before the Geological Society of America "We can indeed for rough purposes, separate petrologists into the "pontiffs" and the "soaks." The pontiff bears the stigma of magma. The magma gives rise to emanations which yield a liquor. The difference between the "pontiff" and the "soak" is that the latter must have his liquor in lavish quantities on all occasions but the former handles his

liquor like a gentleman; he can take it or leave it according to the indications of the individual occasion."

As is the usual case with brilliant scientists, Bowen became recognized as a leader early in his career. In 1912 he entered the Geophysical Labo-



NORMAN L. BOWEN

Recipient of the Roebling Medal of the Mineralogical Society of America

ratory and in 1915 at the age of 28 years he established an international reputation by the publication of "The Later Stages in the Evolution of the Igneous Rocks." In 1928 he developed and expanded this into his book on "The Evolution of the Igneous Rocks," a work which is still a standard and a necessity for all students of this subject. In these works he has expounded the role of fractional crystallization as the major explanation for the diversity of origin of igneous rocks, he has brought for-

ward an overwhelming mass of experimental data developed by himself and his colleagues of the Carnegie Geophysical Laboratory, and has exercised the greatest of ingenuity and skill in showing how the data and principles might apply in support of his thesis. He is recognized as a most doughty opponent in the continuing warfare between certain interpretations based upon laboratory data and conflicting interpretations based on field data.

One of the great ideas developed by Bowen is the significance of the reaction principle in petrogenesis. Pentti Eskola of Finland has called it the most important contribution to petrology in the present century. Bowen has graphically portrayed the principle in a diagram which shows the reaction relation of the mafic minerals as one side of a V and of the plagioclases as the other side of a V, the two sides joining in potash feldspar, muscovite and quartz. This diagram has found application in the relative susceptibility of minerals to weathering and has gone winging through the literature in repeated quotation. I use the term winging deliberately for the reason that the diagram always reminds me of the pattern made by a flock of Canadian geese in their flights during migration.

Other great fundamental contributions have been a quantitative discussion of the heat relationships and the quantitative possibilities of incorporation to be expected where magmas intrude and react with country rock, the succession of mineral assemblages to be expected at successively higher temperatures in the metamorphism of impure carbonate rocks, and in association with Greig the discovery of the mineral mullite as a compound of major importance for the ceramic industry.

One of the great merits of his work and one for which all petrologists are profoundly grateful is the effort which he has made to give examples of the application of the experimental physio-chemical data and principles to petrological problems. In this connection he has diligently sought to visit the classic localities for problems of igneous rocks. He has personally studied the Bushveld of South Africa, the alkalic lavas of East Africa, the peridotites of Skye, the Adirondack and Canadian anorthosites, and there are but few noted petrologic localities in the United States which Bowen has not visited in continuing his own education.

Dr. Bowen has served as Professor of Mineralogy at Queens University (Canada) for 2 years, as Charles L. Hutchinson distinguished service professor of petrology at the University of Chicago for 10 years, and as petrologist with the Carnegie Geophysical Laboratory for 26 years. It is eminently fitting that the award by his own specialistic society should take place in Washington where he has for so long carried on his work.

Dr. Bowen has been the recipient of many distinguished honors includ-

ing the Penrose medal of The Geological Society of America, the Willet G. Miller medal of the Royal Society of Canada, and the Wollaston medal of the Geological Society of London.

An incident may serve to emphasize the modesty of the man to whom today we award the Roebling medal. When the International Geological Congress met in the United States in 1933, I was a member of the trans-continental excursion. One day I casually introduced one of the most distinguished European petrologists to Bowen who was seated in a group in the smoking room of the pullman car. Conversation was general for some time. Shortly after we returned to our regular seats the European petrologist came over to me and in a tone of wonderment and awe said "Was that *the Bowen*, the great petrologist?" I replied, yes that was *the Bowen*, the great petrologist.

Mr. President, it is a personal satisfaction and an honor to present to you "*the Bowen*," a past-president of this society, for the award of the Roebling medal of the Mineralogical Society of America.

PRESENTATION

President George Tunell handed the medal to Dr. N. L. Bowen with the words:

Norman Levi Bowen, in recognition of the new conceptions of the crystallization processes of igneous rocks and the recrystallization processes of metamorphic rocks that you established by precise physico-chemical experiments and field studies in areas of critical exposures, and also in recognition of your determinations of the chemical compositions, stability fields, and physical properties of many minerals, I have the honor to present you with the Roebling Medal of the Mineralogical Society of America.

ACCEPTANCE OF THE ROEBLING MEDAL OF THE MINERALOGICAL SOCIETY OF AMERICA

NORMAN L. BOWEN, *Geophysical Laboratory, Washington, D. C.*

Mr. President, Professor Buddington, Fellows and Members of the Mineralogical Society, and guests:

Yes, it is a great handicap to be insignificant-looking. If I were about 6 feet 5 instead of about 5 feet 6, with a few other improvements too of course, no one would have any doubt that I was a good petrologist, whether I had done any petrology or not. In that connection I think I'll tell you a story, a true story. Some years ago, at meetings just such as these, a friend of mine hurried up to me in the corridor and said, "Oh, there you are. Come with me. Anstruther Perkins (the name is fictitious but indicative) wishes to meet you. Says he has never met Bowen, and I want to be the one to introduce you, so that I may witness his disappointment." It follows me everywhere. Even here, Professor Buddington, while praising me extravagantly, as is his duty on such an occasion, nevertheless manages to suggest, in his sly way, that I am a Canada goose.

Now I am not going to indulge in any display of false modesty and say that the branch of mineral investigation to which I have devoted my life is an unimportant one. I think it is the most important. The problems of the genesis of minerals and rocks are their fundamental problems. I would not go so far as to suggest that the laboratory method of attack on mineral genesis is superior to field attack. They supplement each other, and where they seem to point to different conclusions, the answer is more and better field work and more aptly conceived laboratory work. Whenever opportunity presents, a laboratory investigator should himself get into the field in order that he may have first-hand acquaintance with the problems of nature. As Professor Buddington has stated, I have done some work in the field. I choose an area where a Buddington has been toiling arduously for several years, and get him to show me around, preferably examining outcrops that one can ride right up to in an automobile. When it becomes necessary to ride a horse to an outcrop my prowess is much less marked, as many of my friends can attest, and if it comes to actual foot-slogging, well, least said soonest mended. But to others I always recommend frequent indulgence in this health-giving activity.

Teamwork in science is something of which we hear much nowadays. The visits to Buddingtons which I have mentioned are an example. There is teamwork within laboratories, too. Perhaps the most famous (as some

would say, infamous) example was the Manhattan Project. But teamwork was the hall-mark of the Geophysical Laboratory long before there was any need for a Manhattan Project. Cooperation between physicists, chemists, and geologists was and is the basis of the organization. The Council of the Society is well aware of this, and I am sure had my colleagues in mind, as well as me, in making this award. I would accomplish nothing without my associates. Several of my colleagues, including myself, are very close to the end of our careers as laboratory investigators, but we are building up a stable of prancing young thoroughbreds. See that your field glasses are in order; you will need them to follow these colts.

Scientific research is its own reward. I venture to assert that if the Roebling award were discontinued tomorrow, not a single mineral investigator would cease and desist from his labors on that account, or even slacken his pace. Nevertheless, when such an award as the Roebling Medal comes to one, if my own feelings are any criterion, it is received with great pleasure, mixed with a disturbing sense of unworthiness and a resolve to do better. It is especially gratifying to be introduced by you, Professor Buddington, the leading educator of Canadian geologists, and to receive the medal at your hands, Mr. President, both of you alumni of the Geophysical Laboratory.

Mr. President, I cannot find the words to express my feelings, but I do thank the Mineralogical Society of America most heartily for the great honor it has done me today.

MEMORIAL OF CLARENCE NORMAN FENNER

FRED. E. WRIGHT, (retired) *Geophysical Laboratory, Washington, D. C.*

Clarence Norman Fenner was born on July 19, 1870, on a farm near Paterson, New Jersey. His father, William Griff Fenner, born in 1834 in Chili near Rochester, New York, was the son of a clergyman who, after graduation from Oxford University, migrated with his wife from England to New York state. The son at fifteen years of age left home to make his way in New York City. He was successful and in time became a member of the mercantile firm of Field, Chapman, and Fenner. He bought a farm near Paterson and in 1865 married Miss Elmina Jane Carpenter of North Greece near Rochester, New York. Miss Carpenter was the daughter of Dr. Carpenter, a country physician of English descent. Mr. Fenner commuted each week day between his home and the city. It was in this house that Clarence was born and in which he died on December 24, 1949, 79 years later. In the early part of this century the town of Clifton was extended to include this area which is two miles from Paterson.

Clarence Fenner was educated in private schools in Paterson, namely, that of Miss Riggs, of Mr. Waters, and of Dr. McChesney. In 1888 he spent the summer in Europe in company with Dr. McChesney. On his return home he entered the School of Mines of Columbia University and in 1892 graduated with the degree of Engineer of Mines. He became a mining geologist and saw service in this country, in Canada, in Mexico, and in South America. In 1907, after fifteen years' experience in the field, he returned to Columbia University to earn his Master's degree in 1909 and the Ph.D. in 1910.

Thereupon he joined the staff of the Geophysical Laboratory of the Carnegie Institution of Washington as Petrologist. He remained with the Laboratory until 1938 when he retired, because of age, after 28 years of research work chiefly on petrological and mineralogical problems. He returned to the old home in Clifton to live with his brother Herbert and to continue his researches on petrological problems in his own field; this included a trip to South America to revisit some of the petrologically interesting localities. While in Arequipa, Peru, he prepared a long paper on the geology of the Arequipa area; this paper was published in Spanish by the University of Arequipa after translation by Professor Carlos Nicholson of the University. This article was illustrated by 25 plates.

Dr. Fenner's first independent research problem in the laboratory was the elucidation of the stability relations of the silica minerals. In this field he showed remarkable aptitude and skill in manipulation of laboratory apparatus and for obtaining significant data of observation. He tested



CLARENCE NORMAN FENNER
1870-1949

and retested his conclusions with the result that his data on the silica minerals have stood the test of time and are now accepted. He studied the stability ranges of quartz, including its reversible inversion at about 575°C to the high temperature form which is stable to $870^{\circ}\pm 10^{\circ}\text{C}$. The stable form from this temperature to $1470^{\circ}\pm 10^{\circ}\text{C}$ is β_2 -tridymite. Above this temperature β -cristobalite is stable to its melting temperature at $1713^{\circ}\pm 5^{\circ}$, according to later determinations by J. W. Greig. Fenner found for the two low temperature forms of tridymite: α -tridymite stable below 117° and β_1 -tridymite stable between 117° and 163°C ; both inversions are rapid and reversible. The high temperature form, β_2 -tridymite persists between 163° and $1470^{\circ}\pm 10^{\circ}$ at which temperature it inverts very sluggishly but reversibly to β -cristobalite. This mineral in turn also has a low temperature form, α -cristobalite; the inversion temperature ranges between 220° and 275° depending on the previous heat treatment of the sample; the inversion is rapid and reversible. No satisfactory explanation has yet been found for the observed changes in the inversion temperature.

Fenner's thesis for the Ph.D. degree was on the Watchung basalt and the paragenesis of its zeolites and other secondary minerals. In this thorough study he became intensely interested in the replacement of minerals by others as the result of changes in environment and was able to use this knowledge to advantage later in investigations on replacement minerals produced by pneumatolytic and hydrothermal processes. In his extensive field studies of ore deposits in mines he had been impressed by the magmatic transfer of ore-producing elements either in the gaseous state as pneumatolitic emanations or as hydrothermal solutions. His later studies at Katmai, Alaska, at Yellowstone Park, Wyoming, and elsewhere in North and South America convinced him of the soundness of this hypothesis and led him to reject the hypothesis that fractional crystallization is the principal, if not the sole, factor leading to differentiation in magmas.

In connection with the secondary minerals occurring in the Watchung basalt Fenner solved two problems, namely: (a) the early appearance of albite crystals in the cavities of the basalt; and (b) the former presence of babingtonite crystals which were subsequently removed by solution with only their casts remaining. Fenner discovered in 1914 in the Francisco quarry at Great Notch a small number of babingtonite crystals which had escaped solution and which exhibited the optical and crystallographical properties characteristic of babingtonite. As a rule its crystals occur enveloped in decomposition products; but they were observed so often in cavities that little doubt exists that they are portions of the original crystals. They are found associated with crystals of anhydrite.

The quartz pseudomorphs after babingtonite show sharp prismatic angles which agree fairly well with those of babingtonite. Fenner's recognition of the casts as those of original babingtonite crystals was accepted by mineralogists and thus cleared a mystery that had long puzzled them.

During World War I the Geophysical Laboratory was assigned to aid in the production of optical glass. In December 1917 Dr. Fenner, after some months at the Bausch and Lomb plant in Rochester, was placed in charge of the optical glass plant of the Spencer Lens Company at Hamburg, New York. He succeeded in producing excellent optical glass from the start; he placed the plant on a production basis and extended its capacity many fold. By so doing he contributed his share to the success of his country in winning the war.

In 1919 he was a member of the National Geographic Society's expedition to Mt. Katmai, Alaska; in 1923 he was the leader of the Geophysical Laboratory's expedition to the Katmai region. Between 1928 and 1934 he conducted geological investigations in Yellowstone National Park and became still more interested in magmatic and volcanic processes. At the same time he devoted much energy in developing methods for study of the uranium and thorium minerals and in devising chemical methods for their separation and analysis. These were important factors in studies bearing on the age of the earth and its determination by various methods.

Dr. Fenner's main interest was not in mineralogy nor in chemistry, though he made many excellent analyses of rocks and was well trained in physical chemistry; his interest was chiefly in petrology and in ore deposits, in their development and mutual relationships and in the chemical and mineralogical changes which accompany shifts in environment. His contributions to volcanology, based on extended field studies in the Katmai region, in Yellowstone Park and other areas, followed by painstaking laboratory investigations of the collected field specimens are recognized as fundamental to the development of the science of volcanology. In the Katmai region he proved that the highly siliceous rhyolite magma on rising from the depths encountered near the surface beds of overlying andesite. Materials from the andesite were engulfed and incorporated into the rhyolite, thus forming masses whose composition was intermediate between that of the rhyolite and of the andesite. He proved that this absorption and partial digestion of the andesite by the rhyolite took place near the surface and not at depths; that the sources of energy both for the assimilation and for the violent explosions themselves were the escaping magmatic volatiles whose exothermic chemical reactions produced the necessary heat. He showed that the intermittency in violent volcanic explosions is due to the time required for the exothermic reactions in the

escaping gases to develop. He proved by chemical analyses that in the series of intermediate rocks the variation in the amount of each chemical oxide was linear. This proof by Fenner of the importance of the transfer of material in the gaseous state and of the surficial character of some of the most appalling volcanic outbursts has revived interest in the general subject of pneumatolytic and hydrothermal processes, and called attention to their importance in the crystallization and differentiation of igneous rocks.

Personally Dr. Fenner was quiet and unassuming, thoroughly reliable in every way and ever conscious of our restricted knowledge. In his approach to problems he was strictly scientific and was loath to draw a conclusion before probing its validity from all directions. He was a member of many scientific and honorary societies in this country and abroad.

It was a real pleasure for us of the Laboratory to know him well and to discuss problems with him. His mind was of the rare type, keenly analytical but tolerant and conscious of how little we know of what is to be known. Ever a gentleman and with ability to think and to express himself clearly, he was not dogmatic nor sarcastic even under trying conditions. We knew that in his passing we had lost a good friend and petrology had lost one of its best observers and expounders.

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PROCEEDINGS OF THE THIRTY-FIRST ANNUAL MEETING OF THE MINERALOGICAL SOCIETY OF AMERICA AT WASHINGTON, D. C.

C. S. HURLBUT, JR., *Secretary*

The thirty-first annual meeting of the Society was held on November 16-18, 1950, at the Hotel Statler, Washington, D. C. Scientific sessions were held in the morning and afternoon of November 16 and the morning of November 18, at which thirty-four papers were presented. Five papers were read by title. Five additional papers were presented in the afternoon of November 18 at a Symposium on *Crystal Growth*.

The annual luncheon of the Society on November 16 was attended by 154 fellows, members and guests. Following the luncheon, Arthur F. Buddington introduced the recipient of the Roebling Medal, Norman L. Bowen. Preceding the afternoon Scientific Session on November 16, the retiring President, George Tunell, addressed the Society on *Mineral Formation in Natural Chemical Systems Under Equilibrium and Non-equilibrium Conditions*.

The Council at its meeting on November 15 appointed a committee to consider candidates for nomination to fellowship. This is to be a continuing committee of six members, two new members to be appointed each year to serve for three years. It is hoped that Fellows of the Society will suggest candidates to the committee members.

NOMINATING COMMITTEE FOR FELLOWS 1951

3 years—Samuel Zerfoss and William Parrish

2 years—E. F. Osborn and George Faust, Chairman

1 year—H. W. Fairbairn and C. O. Hutton

REPORT OF THE SECRETARY

To the Council of The Mineralogical Society of America:

ELECTION OF OFFICERS AND FELLOWS

Four hundred and eight ballots were cast in the election of officers: 135 by fellows, 273 by members of the Society. The officers elected to serve in 1951 are:

President: Adolf Pabst, University of California, Berkeley, California.

Vice-President: Michael Fleischer, U. S. Geological Survey, Washington, D. C.

Secretary: C. S. Hurlbut, Jr., Harvard University, Cambridge, Massachusetts.

Treasurer: Earl Ingerson, U. S. Geological Survey, Washington, D. C.

Editor: Walter F. Hunt, University of Michigan, Ann Arbor, Michigan.

Councilor (1951-54): George T. Faust, U. S. Geological Survey, Washington, D. C.

According to the provisions of the Constitution, the following have been elected to fellowship:

J. Dabney Burfoot, Jr., Cornell University, Ithaca, New York.

Alexander de Bretteville, Jr., Signal Corps Laboratories, Fort Monmouth, New Jersey.

Jacob E. de Villiers, Geological Survey, Pretoria, South Africa.

Gabriel Donnay, Johns Hopkins University, Baltimore, Maryland.

Howard Tasker Evans, Jr., Philips Laboratories, Inc., Irvington-on-Hudson, New York.

Robert Bury Ferguson, The University of Manitoba, Winnipeg, Canada.

Rui Ribeiro Franco, University of Sao Paulo, Sao Paulo, Brazil.

Julio Garrido, Lagasca 123, Madrid, Spain.
 Julian R. Goldsmith, University of Chicago, Chicago, Illinois.
 Harold Lloyd James, U. S. Geological Survey, Iron River, Michigan.
 Charles D. Jeffries, Pennsylvania State College, State College, Pennsylvania.
 J. Laurence Kulp, Columbia University, New York, New York.
 Joseph Stevens Lukesh, General Electric Company, Schenectady, New York.
 Charles Mauguin, Sorbonne, Paris, France.
 W. S. Tangier Smith, Menlo Park, California.
 William Hodge Taylor, Cavendish Laboratory, Cambridge, England.
 Robert Mitchell Thompson, University of British Columbia, Vancouver, B. C., Canada.
 Frans Erik Wickman, Swedish Museum of Natural History, Stockholm, Sweden.

CHANGES IN THE CONSTITUTION AND BY-LAWS

The Council of the Society at its meeting in 1949 approved several changes in the Constitution and By-laws of the Society. The complete constitution, incorporating these changes, was printed in the July-August 1950 issue of *The American Mineralogist*. The general membership was asked to accept or reject the constitution as there printed. The changes were accepted with 383 votes in the affirmative, and 5 votes in the negative.

MEMBERSHIP STATISTICS

November 1, 1950

	1949	1950	Gain	Loss
Correspondents	5	5	0	0
Fellows	290	307	19	2
Members	746	747	127	126
Subscribers	630	632	49	47
	1671	1691	195	175

The above figures show a net gain of 17 fellows, 1 member and 2 subscribers. Considering the four groups together there is a total gain of only 20. The present total of members and subscribers is 1691.

The Society lost through death two fellows: Clarence N. Fenner of Clifton, New Jersey, and Martin A. Peacock of the University of Toronto, Canada.

Respectfully submitted,

C. S. HURLBUT, JR., *Secretary*

REPORT OF THE EDITOR FOR 1950

To the Council of the Mineralogical Society of America:

In presenting the annual editorial report at this time, the same policy will be followed as that used a year ago. With five issues published and only the November-December issue in press, sufficient data are now available to give a general survey of the year's activity. When the editor's report appears in print in the March-April number certain data not now available will be incorporated so that the complete printed report can be compared with those given for previous years.

From the standpoint of articles published and total pagination the year 1950 will long be remembered as one of unusual accomplishments. Aside from the normal issues, attention should be called to two so-called special numbers, authorized by the Council, and devoted to specific assignments. The first, sponsored by the Walker's Mineralogical Club continued the series of *Contributions to Canadian Mineralogy*, and appeared as the May-

June issue under the editorship of the late Professor M. A. Peacock. In this number the guest editor assembled 14 interesting papers on varied mineralogical subjects and assumed full responsibility in seeing these contributions through the press.

The issuance this year of a volume of approximately 1100 pages is due in large measure to the greatly enlarged September–October number dedicated to Professor Emeritus Esper S. Larsen, Jr. This number alone carried 32 articles and totaled 344 pages. To the numerous former students and friends of Professor Larsen, who in one way or another made this number possible, we are extremely grateful.

The editor wishes to take this occasion to express his deep appreciation and thanks to Professor Hurlbut for looking after the numerous details connected with the assembling of the manuscripts and the printing of this special issue.

The 1100 page volume of the current year represents an increase of approximately 200 pages over last year and represents the largest volume in the history of the Society with the single exception of the volume of 1937 that included the special Palache number.

While there has been some delay this year in the appearance of certain manuscripts due to the special issues in which the articles were restricted to specific assignments, the circumstances which have caused the delay this year are not likely to occur again in the near future, so that our normal publication schedule can again be established in 1951.

While it is impossible at this early date to state with any degree of accuracy the printing demands that will be made on the Journal in 1951, still it would seem that the volume will not reach the size of that produced this year. At present no special enlarged issues are contemplated and a normal volume of about 850 to 900 pages might reasonably be stated as a likely forecast.

In regard to the *Contributions to Canadian Mineralogy*, the late Professor Peacock wrote some time ago that he hoped that a number in 1951 could be arranged and desired to have this item brought to the attention of the Council for their consideration and approval. In addition some 25 papers of varying lengths are now on hand, so that the 1951 volume would likely assume the proportions of the volume of a year ago.

Financial support this year, aside from the income obtained from dues and interest received from the Society's investments, has come from two sources. The sponsors of the Harvard number have assisted in defraying the excessive cost of the enlarged September–October number and the Geological Society of America has continued its generous assistance in defraying a substantial portion of the ordinary printing costs. For this support the Society is indeed very grateful as otherwise dues would have to be increased or restrictive measures applied to keep the costs within our established income. Reduction in the length of manuscripts through condensation would go a long ways to help ease the financial situation as printing costs are not likely to decline, but unfortunately condensation is the hardest commodity to sell to the author who naturally desires full and what he considers adequate coverage.

The current volume as indicated above will run about 1100 pages and contain 92 articles. Seventeen additional articles appearing under Notes and News give an over-all total of 109 published manuscripts for the year (last year it was 73 and 25, totaling 98). Due to the widespread source material secured for the Larsen number, no less than 32 contributions from 10 countries were received from contributors residing outside of the States; 17 of which came from Canada.

These contributions were received from 122 contributors associated with 61 different Universities, research bureaus, and technical laboratories. Two new minerals were described in detail for the first time: hühnerkobelite and wherryite. The accompanying Table 1 summarizes in detail the distribution of subject matter in Volume 35.

Respectfully submitted

WALTER F. HUNT, *Editor*

TABLE 1. DISTRIBUTION OF SUBJECT MATTER IN VOLUME 35

Subjects	Articles	Pages	Per Cent of Total
Leading articles*			
Descriptive mineralogy.....	22		
Chemical mineralogy.....	23		
Structural crystallography.....	16		
Petrography.....	12		
Geometrical crystallography.....	5		
Optical crystallography.....	4		
Mineralography.....	1		
Memorials.....	2		
Miscellaneous.....	7		
	92	950.....	86.8
Shorter articles.....	17	36}	
Notes and News.....	13	7}	
Proceedings of Societies.....	6	73}	13.2
Book reviews.....	18	18}	
New mineral names.....	30	10}	
	176	1094	100.0
Illustrations.....	312		
Index, Title page, Table of contents.....		20	
Grand total.....		1114	

* Leading articles average 10.3 printed pages each.

REPORT OF THE TREASURER FOR 1950

To the Council of the Mineralogical Society of America:

Your treasurer submits herewith his annual report for the year beginning November 1, 1949 and ending October 31, 1950.

RECEIPTS

Dues and subscriptions.....	\$ 7,658.48
Sale of back numbers.....	1,305.41
Authors' charges on reprints.....	1,217.90
Interest and dividends from endowment.....	3,725.20
Geological Society of America aid for printing the Journal.....	5,299.34
Advertising.....	599.16
Sale of Index, volumes 21-30.....	93.90
Sale of Index, volumes 1-20.....	7.70
	\$19,907.09
Cash on hand, November 1, 1949.....	3,601.54
	\$23,508.63

DISBURSEMENTS

Printing and distribution of the Journal (6 issues)	\$11,346.15
Printing and distribution of separates	1,377.15
To the Editor, Secretary, and Treasurer	1,250.00
Clerical help	550.50
Postage and express	185.10
Printing and stationery	77.06
Office equipment and supplies	117.37
Roebling Medal	139.59
New securities purchased	696.00
1949 Program and abstracts	293.63
Expenses of officers to 1949 annual meeting	204.22
Committee expenses	2.83
Safety deposit box	7.80
Back numbers purchased	14.00
Telephone and telegraph	3.32
Checks returned	47.90
Refunds	22.80
Exchange charges on checks	1.15
	<hr/>
	\$16,336.57
Cash balance, October 31, 1950	7,172.06
	<hr/>
	\$23,508.63

The endowment funds of the Society as of October 31, 1950, consist of the following securities:

BONDS

6M Atlantic Coast Line, 4½	\$5,257.50
5M New York Central, 5	4,300.00
5M Southern Railway, 5	5,743.75
4M Cleveland Union Terminal, 5	4,055.00
4C Great Northern, 5½	400.00
	<hr/>
	\$19,756.25

PREFERRED STOCKS

200 shares, Southern California Edison, 4.88	\$5,250.00
100 shares, Union Pacific, 4	4,570.25
60 shares, Jones and Laughlin, A, 5	4,987.50
55 shares, United States Steel, 7	6,946.20
50 shares, Virginia Electric & Power Co., 5	5,942.50
24 shares, Public Service Electric & Gas Co.	728.40
10 shares, Consolidated Edison, 5	1,066.64
	<hr/>
	\$29,491.49

COMMON STOCKS

288 shares, Potomac Electric Power Co.....	\$3,871.00
60 shares, United Fruit Co.....	3,067.50
50 shares, Chesapeake and Ohio Railway.....	2,368.75
50 shares, Pennsylvania Railroad.....	1,468.75
41 shares, American Telephone and Telegraph.....	5,599.32
40 shares, Kroger Co.....	1,990.00
28 shares, Standard Oil of New Jersey.....	1,444.84
1 share, New York, Chicago & St. Louis Railroad.....	37.00
	<hr/>
	\$19,847.16
	<hr/>
	\$69,094.90
Respectfully submitted,	
EARL INGERSO, <i>Treasurer</i>	

DANA FUND

Disbursements are made to needy mineralogists in war areas and to needy families of deceased mineralogists in war areas.

RECEIPTS

Available balance, November 1, 1949.....	\$340.84
Interest.....	1.31
	<hr/>
	\$342.15

DISBURSEMENTS

Disbursed.....	\$129.40
Available balance, November 1, 1950.....	212.75
	<hr/>
	\$342.15
Respectfully submitted,	
EARL INGERSO, <i>Treasurer</i>	

REPORT OF THE AUDITING COMMITTEE

To the President of the Mineralogical Society of America:

The Auditing Committee has examined and verified the accounts of the Treasurer of the Mineralogical Society of America for the fiscal year ending October 31, 1950. The securities listed in the Treasurer's report, with all future coupons on the coupon bonds attached, are in the safety deposit box at the West End Branch of the Washington Loan and Trust Company in Washington, D. C.

Respectfully submitted,
WILLIAM D. JOHNSTON, JR.
WALDEMAR T. SCHALLER
FELIX CHAYES, *Chairman*

ABSTRACTS OF PAPERS PRESENTED AT THE THIRTY-FIRST
ANNUAL MEETING OF THE MINERALOGICAL
SOCIETY OF AMERICA AT WASHINGTON, D.C.
NOVEMBER 16-18, 1950

PETROGRAPHY OF THE WILCOX FORMATION OF ARKANSAS

VICTOR T. ALLEN

Saint Louis University, St. Louis, Missouri

The mineral composition of representative clays and sandstones of the Wilcox formation of Arkansas was determined petrographically in cooperation with the Division of Geology, Arkansas Resources and Development Commission. Samples representing the exposed thickness near Texarkana, Hope, Malvern and Little Rock were studied and show little variation in mineral composition with stratigraphic position or along the strike of the outcrops for the 140 miles from Pulaski County to Miller County. Representative samples of the clays used in ceramics were analyzed chemically and their mineral composition was checked by *x*-ray and differential thermal methods. The principal minerals are kaolinite, quartz and montmorillonite. Small amounts of muscovite, microcline, zircon, tourmaline, rutile, epidote, ilmenite and leucoxene are present in most samples. A few grains of kyanite similar to that which occurs in the schists of North Carolina are present in some samples and suggest that part of the sediment was contributed from Appalachia. The mineral assemblage indicates thorough weathering of the source rock prior to deposition of the sediments. The kaolin minerals were formed by long continued leaching by neutral or slightly acid groundwaters under favorable climatic and physiographic conditions. The montmorillonite was formed from suitable source rocks by alkaline solutions under conditions of poor drainage. The clay minerals were eroded, mixed with quartz and other minerals and deposited in the sea which occupied this area during Eocene time.

THE SHONKIN SAG LACCOLITH REVISITED

JULIAN D. BARKSDALE

University of Washington, Seattle, Washington

Re-study of the earlier collected data and re-examination in the field has convinced the author that the gross layering in the Shonkin Sag laccolith is due to differentiation in place from one and possibly two closely connected intrusions of shonkinitic magma. The layer exhibiting exotic crystallization of biotite-augite-sanidine is a syenite-pegmatite formed during the early pneumatolytic stage of crystallization. The wider of the hornblende-aegirine syenite dikelets, which cut the pegmatite, represents the consolidation of the magmatic residuum; the narrower ones are the result of solution replacement along cracks. The many sharp contacts between the various rock types in the laccolith are thought to be due to auto-injection caused by tectonic movements which involved the laccolithic disc while it was solidifying and which continued after it had cooled.

**THE MORPHOLOGY AND STRUCTURE OF THE SERPENTINE MINERALS
ANTIGORITE AND CHRYSOTILE**

THOMAS F. BATES AND JOHN F. MINK

Pennsylvania State College, State College, Pennsylvania

Studies with the electron microscope supported by *x*-ray and DTA data show that natural and synthetic chrysotile are composed of tube-like fibers, whereas antigorite con-

sists of plates and laths. In both instances the morphological characteristics of the artificial materials are better defined than those of the natural minerals. The units of chrysotile always show curvature of the (001) plane parallel to the *a* or *b* axis. In antigorite this curvature is less marked or absent entirely.

Interpretation of the data suggests that antigorite and chrysotile differ morphologically only in the degree of lateral curvature affecting their component units. Evidence indicates that this curvature is most pronounced where the ideal serpentine composition is obtained, such as in synthetic chrysotile, and decreases with the amount of trivalent ions replacing magnesium in the structure. It is believed that the degree of curvature depends upon (1) the discrepancy in dimensions of the "sheets" which make up the unit of structure, and (2) the strength of the bonds between the units. This is in accord with the theory previously proposed to account for tubular crystals of endellite.

PHOTOGRAPHS OF THE ATOMS IN THE STRUCTURES OF MINERALS

M. J. BUERGER

Massachusetts Institute of Technology, Cambridge, Massachusetts

A generalized microscope has been constructed which permits such magnification that the atoms in crystal structures can be seen and photographed. In this microscope light of short wavelength is used as far as the first diffraction image, and then light of a longer wavelength is used from this diffraction image to the final image. For such a microscope the magnification contains not only the usual factor of the image to object ratio, but also a second factor consisting of the ratio of the wavelengths of the second to the first kind of radiation. When visible light and x-rays are used for these radiations, this second factor is about 10,000 and the whole magnification is about 300,000 diameters. This is sufficient to enable one to just see the individual atoms of a crystal structure. Unfortunately, a loss of phase occurs when the wavelength is changed. This is made up by the use of phase shifters composed of mica of uniform thickness. The apparatus actually used for this generalized kind of microscopy is described, and photographs are shown of the crystal structures of pyrite, marcasite, and KH_2PO_4 .

CORUNDUM DEPOSITS OF MONTANA*

STEPHEN E. CLABAUGH

University of Texas, Austin, Texas

No other part of North America presents such a rich variety of corundum deposits as central and southwestern Montana. Multi-colored sapphires have been recovered from placers near Helena, Philipsburg, and Deer Lodge, chiefly for use as watch jewels. The Yogo sapphire mine in Fergus County was one of the few important sources of precious stones in the United States and the only deposit in the world where sapphires were recovered directly from igneous rock. Deposits of ordinary corundum in metamorphic rocks in Gallatin and Madison Counties have been investigated as possible sources of natural abrasive, but they were found to be small and low in corundum content.

The placer sapphires are chiefly abraded short prismatic crystals of pale color, which probably were derived from andesitic (?) dikes similar to the dike exposed at French Bar on the Missouri River. At Yogo Gulch the sapphires are platy to equidimensional pyramidal crystals of excellent blue color in an altered analcine-bearing lamprophyre. Reaction of the lamprophyric magma with kyanite-bearing inclusions may have caused precipitation of

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corundum at an early stage in the cooling of the rock. Later the sapphires were partly dissolved and coated with spinel.

The abrasive corundum deposits are concordant tabular to lenticular bodies enclosed in metamorphosed pre-Cambrian sedimentary rocks. Feldspar, mica, sillimanite, and rutile accompany the corundum, and the texture of the rock ranges from fine-grained and schistose to pegmatitic. These deposits have recently been described as corundum pegmatites, but their form, distribution, and composition suggest an origin by metamorphism of alumina-rich sedimentary lenses.

LUMINESCENCE ACTIVATION OF ZEOLITE MINERALS BY BASE EXCHANGE

ESTHER W. CLAFFY AND JAMES H. SCHULMAN

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Natural zeolite minerals do not luminesce, but luminescence may be artificially induced by substituting an activator such as Mn^{++} , Pb^{++} , Ag^+ , or Cu^{++} for part of the minerals' cations (usually Na^+ and/or Ca^{++}) through base exchange. The powdered mineral is merely soaked in dilute aqueous solution of the activator metal (such as 1×10^{-3} M Cu^{++} solution) at room temperature for as little as 15 minutes. Only $2 \times 10^{-4}\%$ (wt.) Cu in stilbite produces luminescence. Luminescence activation is a more sensitive test for base exchange than optical or x-ray diffraction properties, which are altered only after extensive base exchange.

Base-exchanged Cu-zeolites and Ag-zeolites show green and blue cathodeluminescence (under spark coil discharge or cathode ray excitation), respectively after heating at 300° C. (dehydrating, to an undetermined extent), some Cu- and Ag-zeolites also develop photoluminescence under ultraviolet excitation (2537Å, 3650Å). Mn-, Pb-, and (Mn+Pb)-zeolites, which in their normal hydrated state are unresponsive to any excitation source, also develop cathodeluminescence and/or photoluminescence after being dehydrated. When these dehydrated, base-exchanged zeolites are rehydrated, they lose their luminescence.

Luminescence of the zeolites is not a surface adsorption phenomenon, but is related to the entry of the activator ion into the crystal lattice. Removal of the activator ion from the crystal lattice by a reversed base exchange process eliminates the luminescence. Development of luminescence in base-exchanged zeolites is also a reversible process dependent on the state of hydration or dehydration. The openness of the zeolite crystal lattice and its structural water are significant. All three structure-types of zeolites have been investigated: the fibrous (natrolite), lamellar (stilbite, heulandite), and rugged tri-dimensional (chabazite).

ISOMORPHISM OF SYNTHETIC FLUORINE-AMPHIBOLES

J. E. COMEFORO, R. A. HATCH, AND WILHELM EITEL

U. S. Bureau of Mines, Electrotechnical Laboratory, Norris, Tennessee

In the course of research on the synthesis of asbestos, experimentation was conducted on the crystallization of amphiboles from melts containing fluorine. Major emphasis was given to the study of fluor-tremolite ($Ca_2Mg_5Si_8O_{22}F_2$) and fluo-richterite ($Na_2CaMg_5Si_8O_{22}F_2$). X-ray, chemical, and microscopic analyses indicated a number of isomorphous replacements in the structure by elements that are not observed in natural amphiboles.

The general formula for the complex amphibole group is represented by $(WX)_{2-3}Y_5(Z_4O_{11})_2$. The positions of the W ions in $(WO_4)_2$ coordination were filled completely by Na^+ , Li^+ , and Ca^{++} . Attempts to substitute K^+ resulted in the crystallization of mica to the ex-

clusion of amphibole. The positions of X ions in (XO_6) coordination were filled completely by Ca^{++} , Cd^{++} , and partly by Sr^{++} , Ba^{++} , and Na^+ . Magnesium ions could not be substituted in 8-fold coordination. The Y, or (YO_6) positions were satisfied completely by Mg^{++} and partly by Fe^{++} , Zn^{++} , Co^{++} , Ni^{++} , Li^+ , and Al^{+++} . In the tetrahedral group $(ZO_4)B^{+++}$ is capable of occupying at least 25 per cent of the 4-fold positions, Be^{++} 12 per cent, and Cr, V, and Al^{+++} undetermined amounts. It was not possible to replace Si^{+4} by Ge^{+4} . This substitution results in the crystallization of Ge-diopside. Attempts to replace F^- by Cl^- in the fluoramphibole structure by crystallization from melts were unsuccessful.

It was concluded that the extent of isomorphism of fluoramphiboles is more restricted than in fluor-micas.

The field of stability of fluor-amphiboles is small; consequently, the compositional changes brought about by attempted isomorphous substitutions frequently results in the crystallization of olivine, pyroxene, feldspar, and mica. This conforms with natural magmatic differentiation of phases by crystallization.

VARIATIONS IN CHEMICAL COMPOSITION ACROSS IGNEOUS CONTACTS

WILLIAM H. DENNEN

Massachusetts Institute of Technology, Cambridge, Massachusetts

The contact effects resulting from the intrusion of igneous material into various wall rocks are linear compositional changes only in special instances. Large variations in chemical composition from point to point may be present in both intrusive and wall rocks.

A study of these variations by the analysis of closely spaced samples with quantitative spectrographic methods indicates that all of the major rock constituents studied (Si, Al, Fe, Mg, Mn, Ca, Na, and K) are mobile when moderate temperatures and concentration gradients are present. Migration of material is believed to take place late in the cooling history by diffusion along mineral boundaries. The compositional variations resulting from this transfusive migration of hydrated ions are often sufficiently large to mask any compositional changes due to earlier reactions.

The chemical variations found can be related to the original compositions of the wall rock and intrusive material, and to the cooling history of the system. No variations in chemical composition are found across contacts between chemically identical rocks. Generally smooth and continuous variations occur across contacts between rocks of different original composition which have been held at high temperatures. Highly irregular variations are found when the intrusion and wall rock were compositionally different and steep thermal gradients existed. These latter variations may show geochemical culminations and depressions, are wave-like in form, and die out in both directions from the contact zone.

STRATIGRAPHY AND METAMORPHIC RECONSTITUTION OF PARTS OF THE GRENVILLE SERIES IN THE NORTHWEST ADIRONDACKS: A REPORT OF PROGRESS

A. E. J. ENGEL AND CELESTE ENGEL

California Institute of Technology, Pasadena, California

Although most of the metasedimentary rocks of the Grenville series in the northwest Adirondacks have been profoundly reconstituted, stratigraphic relicts exist which enable a coherent, predictable succession to be reconstructed. The pre-metamorphic metasedimentary sequence in the Balmat-Edwards region was perhaps 9,300 feet thick; 15 relict units are identifiable at points along the lithologic trend for 10 or more miles and many smaller subdivisions are recognizable locally. Stable-shelf facies consisting of thin quartzose beds in thicker dolomite zones and a siliceous, sodic shale (tuff?) predominate.

Reconstitution of some dolomite units involved large-scale dedolomitization. The mag-

nesium was widely redistributed and was fixed in assemblages of quartz-diopside-serpentine, tremolite-anthophyllite-serpentine-talc, and granitized hornblende-andesine amphibolites. Variations of the above assemblages may include scapolite, phlogopite, chlorite, calcite, feldspar, hexagonite-tourmaline, anhydrite, hydrogen-sulfide, and graphite. Important constituents added to and irregularly distributed through the marble include alkalies, manganese, fluorine, chlorine, titanium, phosphorus, silicon, water, and locally zinc, lead, and iron sulfides. Calcium and carbon dioxide, especially, were removed.

The thick shale (tuff?) unit was commonly altered to a quartz-biotite-sodic andesine-almandite-microcline migmatite that locally contains sillimanite-bearing facies. Other, granitized portions were progressively enriched in alkalies and depleted in iron, magnesium, and calcium; these granitic rocks include gneissoid-augen, equigranular-alaskitic, granulitic, and quartz-syenitic types.

Many bodies of the reconstituted rocks tend to follow the original bedding, but other bodies are irregular and blur or obliterate the bedding.

Although of regional scope, the reconstitution was an intricate composite of "contact" and "regional" metamorphism.

CHALCANTHITE BY THE X-RAY PRECESSION TECHNIQUE

D. JEROME FISHER

University of Chicago, Chicago, Illinois

This paper outlines the *x*-ray precession technique as applied to determining the geometry of the lattice of a triclinic crystal. While the orientation of a triclinic subhedron is less simple for the precession technique than for the Weissenberg, once it is secured the angular constants are much more easily obtained and are far more accurate. The necessary three angular and three linear constants are given from photographs made from a single setting; the results are compared with previous goniometric and *x*-ray studies. The 1-level precession photograph is similar to the Goldschmidt gnomonogram except for scale; this will be illustrated by 0- and 1-level pictures taken on a single film.

SEDIMENTARY AMPHIBOLITES IN THE VAN HORN MOUNTAINS, TEXAS

PETER T. FLAWN

University of Texas, Austin, Texas

Amphibolite, biotite amphibolite, epidote-biotite amphibolite, biotite-anthophyllite amphibolite, anthophyllite amphibolite, almandine amphibolite, epidote amphibolite, and epidosite are found in the Mica Mine area of the Van Horn Mountains, Texas. The amphibolites occur with biotite schists in thin beds and lenses in a thick pre-Cambrian feldspathic quartzite-muscovite schist sequence. Distribution, varied composition, layered character, associated rocks, and transition rocks demonstrate a sedimentary origin. These rocks were originally ferruginous dolomitic marls and ferruginous shales in a sandstone and shale sequence.

Analyses of representative rocks are plotted on ACF diagrams. With low Na₂O, high Fe₂O₃, and excess CaO, epidote takes the place of anorthite. Almandine forms in rocks with excess Al₂O₃ if MgO and FeO act as separate components. With a deficiency of Al₂O₃, ferro-anthophyllite forms instead of almandine. With available K₂O and Al₂O₃, MgO and FeO may act as two components to form biotite and almandine or as one component to form biotite and (when K₂O and Al₂O₃ are exhausted) ferro-anthophyllite.

On Vogt's ACF diagram the rocks fall in the "sedimentary series," corroborating field evidence of sedimentary origin.

SURFACE FEATURES OF CHERT AS STUDIED BY THE ELECTRON MICROSCOPE

ROBERT L. FOLK AND CHARLES EDWARD WEAVER
 Pennsylvania State College, State College, Pennsylvania

Electron microscope study of fracture surfaces of chert, using the replica technique, has shown that at least two and possibly three types of surfaces are present. Specimens which under the light microscope consist of microcrystalline quartz (minute, equant grains), show sharply defined polyhedral blocks with smooth, slightly curved faces—exactly like the shapes occupied by the air cells in soap froth. Arkansas novaculite, a very homogeneous type of chert, possesses this type of surface to a marked degree. Specimens which under the light microscope are composed of chalcedonic quartz (radiating fibers), have a more or less spongy surface, with no evidence of fibrous character when examined with the electron microscope. The sponginess is caused by the presence of abundant spherical water-filled bubbles, all very nearly 0.1 micron in diameter. These bubbles impart a brownish color to the chalcedonic quartz as seen in the light microscope, and decrease the density and refractive index (due to the Christiansen effect), in proportion to their abundance. Chalcedonic quartz without bubbles has the same refractive index as normal quartz. The properties of chert and chalcedony are adequately explained by the hypothesis that it consists of fine-grained quartz plus a variable quantity of free water in the form of distinct bubbles; there is no evidence of admixed opal.

THE IMPORTANCE OF EXCHANGEABLE MAGNESIUM IN THE STUDY OF MONTMORILLONITIC CLAYS*

MARGARET D. FOSTER
 U. S. Geological Survey, Washington, D. C.

In calculating the formulas of members of the montmorillonite group all the magnesium is generally assigned to the Al layer. However, determination of exchangeable cations in 16 montmorillonitic clays, 12 of them the identical specimens for which analyses were quoted by Ross and Hendricks (U.S.G.S. Prof. Paper 205-B), showed that in all of them some of the magnesium was exchangeable, the amount ranging from 0.11 to 1.62 per cent. In two samples exchangeable magnesium amounted to about $\frac{1}{3}$ of the total magnesium. These results indicate that the presence of exchangeable magnesium in the montmorillonitic clays is more common than is generally recognized.

Ross and Hendricks, assigning all magnesium to the Al layer in calculating the formula of montmorillonites, found that the number of ions in octahedral coordination ranged from 2.02 to 2.24. However, a recalculation of the formulae of some of these samples on which exchangeable cations were determined showed that, when total magnesium is corrected for exchangeable magnesium and only nonexchangeable magnesium is assigned to the Al layer and when the determined rather than an average base exchange capacity is used, the number of cations in octahedral coordination does not exceed 2.02, even in those samples in which the number of cations is as much as 2.13 when all the magnesium is assigned to the Al layer. Apparently, in montmorillonite then, substitution of Mg for Al in the Al layer is ion for ion, with no extra ions added in an effort toward internal compensation of charge.

HIGH-TEMPERATURE X-RAY STUDY OF THE POLYMORPHISM OF MgSiO_3

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An investigation of the polymorphic inversions of magnesium metasilicate (MgSiO_3) has been made with the aid of a high-temperature x-ray spectrometer furnace. This study

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which involved the use of both natural and synthetic material, has yielded results which differ considerably from those of previous investigators. On the basis of these results, a new scheme of relations between the three crystalline forms of MgSiO_3 —enstatite, protoenstatite, and clinoenstatite—is proposed.

SYNTHESIS OF STAR SAPPHIRES AND RUBIES

CLIFFORD FRONDEL

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The properties of synthetic star sapphires and rubies manufactured by the Linde Air Products Company are described. The asterism is produced by the exsolution in oriented position of rutile needles from an initially homogeneous solid solution of titanium in corundum single-crystals grown by the Verneuil process. The initial solid solution contains about 0.06 to 0.2 atomic per cent of Ti^{3+} in substitution for Al, together with about 1.4 per cent Cr^{3+} and about 0.3 per cent Fe^{3+} as pigmenting material in ruby and sapphire, respectively. The clear, homogeneous boules are annealed at about 1900°C . to remove strain and are then heated for 2 to 72 hours in an oxidizing atmosphere between 1500° and 1100° to induce precipitation of TiO_2 . The length of the precipitation treatment increases with decreasing temperature. Precipitation does not occur over about 1500° , and exsolved rutile can be dissolved by heating to somewhat over 1500° . The crystal chemistry of the precipitation is discussed, and criteria for the recognition of synthetic asteriated corundum are indicated.

THE CRYSTAL STRUCTURE OF CLAUDETITE (MONOCLINIC As_2O_3)

ALFRED J. FRUEH, JR.

University of Chicago, Chicago Illinois

The unit cell of claudetite has been determined by M. J. Buerger to be $a=5.25 \text{ \AA}$; $b=12.87 \text{ \AA}$; $c=4.54 \text{ \AA}$; $\beta=93^\circ 49'$ and the space group determined to be $\text{P}2_1/n$. The cell contains $4\text{As}_2\text{O}_3$. Due to the darkening of the general radiation streak on the c -axis zero level Weissenberg in the vicinity of the (030) reflection, doubt was cast on the existence of the two-fold screw axis (2_1). The presence of the 2_1 was confirmed by the comparison of the Harker-Patterson syntheses ($\text{P}xoz$) and $\text{P}(x\frac{1}{2}z)$. From these syntheses and from Patterson projections on (001) and (100) the eight arsenic atoms were found to lie in two general four-fold positions. One set at $x=.27$, $y=.10$, $z=.03$ and its symmetry equivalents and the other set at $x=.35$, $y=.35$, $z=0.00$ and its symmetry equivalents. Electron density projections were constructed from those reflections whose phases were unequivocally determined by the location of the arsenic atoms. From these projections the locations of the oxygen atoms could be roughly determined. These are being refined by further electron density projections and intensity checks.

FORMATION TEMPERATURES OF SOUTHERN ILLINOIS BEDDED FLUORITE AS DETERMINED FROM FLUID INCLUSIONS

ROBERT M. GROGAN AND RAYMOND S. SHRODE*

State Geological Survey, Urbana, Illinois

Fluid inclusions in fluorite crystals were studied principally to determine the range of temperature of formation of individual crystals. Measurements were made by heating

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specimens and recording temperatures at which vapor bubbles within the inclusions disappeared. Overall accuracy, excluding pressure corrections, was of the order of 2.5°C .

Some of the fluid inclusions are arranged in geometrically regular planes parallel to cube faces. These are apparently of primary origin. Others form bands along former fractures which approximately parallel the cleavage directions and which cut across each other and the primary planes alike. Historically such inclusions have been called "secondary" although "subsequent" is considered a more meaningful designation. Most of the primary inclusions are smoothly rounded and contain petroleum, whereas "subsequent" inclusions range from jagged to smooth in outline and contain principally a clear fluid.

From measurements on primary inclusions, temperatures at which the crystals grew generally ranged from 85° to 120°C ., with a mean around 100°C . Commonly the indicated growth temperature increased slightly for some distance outward from the crystal center and then dropped off toward the exterior. Measurements on "subsequent" inclusions gave results as high as 185°C ., generally higher than those shown by primary inclusions.

Incomplete bulk analyses of the fluid from inclusions, probably mostly of the "subsequent" variety, show a preponderance of Ca, Na, and Cl ions, and a total concentration of dissolved material amounting to 45,000 parts per million.

SYNTHESIS OF FLUORINE-MICAS AND RELATED LAYER STRUCTURE COMPOUNDS

R. A. HATCH, WILHELM EITEL, AND R. A. HUMPHREY

U. S. Bureau of Mines, Electrotechnical Laboratory, Norris, Tennessee, in Cooperation with Office of Naval Research, Bureau of Ships, and Army Signal Corps

The synthesis of F-micas and related layer structure compounds has progressed far enough to furnish much information on the stability field of these important rock-forming minerals. However, much remains to be done.

Techniques of synthesis used in these investigations involve (1) fusion and crystallization, or (2) solid state reaction. Many compounds such as F-muscovite can be formed only by the latter method. Special techniques often are needed to produce the desired reaction, especially at temperatures below 700°C . Examples of special techniques include the preparation and use of highly reactive batch materials, and the use of special atmospheres. Additionally, the proper choice of batch materials often is vital in obtaining the desired reaction product because in many cases, if not most, equilibrium conditions are not attained during the experiment.

Compounds synthesized include more than 100 different varieties of F-micas. Most of those related to the F-phlogopite subfamily can be crystallized from a melt; however, a few may be formed only by solid state reaction. A few of the F-phlogopites melt congruently but most of them dissociate below the melting temperature. All members of the F-muscovite and related lithium micas melt incongruently and can be formed only by solid state reaction. The synthesis of F-talc and F-pyrophyllite has not yet been achieved although an interesting F-pyrophyllite has been made. A compound which seems to be closely related to the natural clay mineral, hectorite, has been prepared from dry melts.

ORIGIN OF GRANOPHYRE IN DIABASE IN SOUTHEASTERN PENNSYLVANIA*

PRESTON E. HOTZ

U. S. Geological Survey, Washington, D. C.

Small bodies of granophyre occur in the upper part of Triassic diabase intrusions in southeastern Pennsylvania. Near Harrisburg granophyre in the upper part of a diabase

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sheet was penetrated in a diamond drill hole. Drill core specimens show a gradational sequence from diabase to granophyre.

New data include 10 new chemical analyses and the results of petrographic study of a series of specimens from the drill core. The sequence, from diabase to granophyre, shows progressive increase in alkalis and silica; iron increases into transitional granophyric diabase, then decreases in the granophyre. The chemical variations are accompanied by changes in mineralogy.

It is concluded that crystal differentiation in a large sheetlike intrusion most satisfactorily explains the genesis of granophyre in this region. Prior to complete solidification, a residual liquid rich in iron, alkalis, silica, and volatiles accumulated locally in the upper part of the diabase sheet. In some places volatile-rich iron-bearing solutions that carried magnetite escaped into the overlying sedimentary rocks; the remaining liquid crystallized to fine-grained granophyre.

A MONOCHROMETER UTILIZING THE ROTARY POWER OF QUARTZ

CORNELIUS S. HURLBUT, JR.

Harvard University, Cambridge, Massachusetts

The Emmons method of double variation for the determination of refractive index would be used more extensively if a more adequate light source were available. It is believed that this instrument meets the need.

The instrument utilizes the dispersion produced by the rotary power of quartz to obtain essentially monochromatic light. Four basal sections of quartz are used. Number 1 is of left-hand quartz 60 millimeters thick. Numbers 2, 3, and 4 are of right-hand quartz 30, 15, and 7.5 millimeters thick respectively. The sections are arranged so that a columnated and polarized light beam will pass through section 1 into section 2, 2 into 3, and 3 into 4. The light on emerging from section 1 passes through a polaroid plate, which eliminates nine bands equally spaced through the visible spectrum but permits nine others to pass into section 2. As the light emerges successively from each of the sections, it passes through polaroid discs which eliminate half of the number of bands that entered. When it has passed through section 4 and its polaroid, all but one band with an average width of 150 Å has been effectively eliminated.

By rotating simultaneously all four of the quartz sections with their attached polaroids, it is possible to change continuously the position of the single pass band so that it will move completely through the visible spectrum.

Because of the large aperture of the instrument, adequate light of all wave lengths can be obtained by using a tungsten filament lamp as a light source.

GROWTH OF CRYSTALS OF MICA AND RELATED FLUOSILICATES

HERBERT INSLEY

National Bureau of Standards, Washington, D. C.

It is well known that fluorine may be substituted, in whole or in part, for hydroxyl in many crystalline silicates and chemically related compounds without essential structural changes. The structural and compositional limitations to such substitutions are considered with particular respect to bonding and to geometrical configuration. Possibilities of synthesis of such fluorine substituted minerals as mica amphibole, chrysotile, topaz, apatite, and clay minerals are reviewed. Some principles are stated regarding the control of orientation in synthesis of platy and fibrous fluorine minerals by temperature gradients and other methods.

GEOLOGICAL SIGNIFICANCE OF VARIATIONS IN THE HIGH-LOW INVERSION OF QUARTZ

M. L. KEITH AND O. P. TUTTLE

Geophysical Laboratory, Washington, D. C.

Investigation of variations of the high-low inversion of quartz, has been refined and extended to include almost 300 samples of synthetic and natural quartz from various sources.

The inversion temperature of samples of quartz grown in the laboratory under controlled conditions of pressure and chemical environment is in general an inverse function of the temperature of growth. That simple relationship is complicated however, by the presence of an element such as germanium, which can substitute for silicon in the comparatively "tight" quartz structure, with profound effect on the inversion.

On the basis of temperature of inversion, natural quartz samples fall into fairly well-defined groups which are clearly related to the conditions of origin. Quartz from rhyolites and related intrusive porphyries and welded tuffs, inverts at temperatures clustered around 572.7° . Quartz from granites, with a few exceptions among those studied, inverts at higher temperatures clustered around 573.2° . "Granite" quartz and "rhyolite" quartz are completely separated on a plot of heating inversion versus cooling inversion. Most samples of vein quartz give high inversions, while samples of quartz from pegmatites show wide variations, even within one pegmatite mass. Details are presented regarding the inversion of quartz from zoned pegmatites and zoned crystals. Some suggestions are made as to the possible significance of the inversion characteristics of quartz from different types of granite.

ORIGIN OF HORNBLende RIMS ON ADIRONDACK GARNET

S. BENEDICT LEVIN

Signal Corps, Engineering Laboratories, Fort Monmouth, New Jersey

Rims (shells) of coarse hornblende surround large garnet crystals in coarse metagabbro at the Barton garnet deposit and other occurrences of this type in the southeastern Adirondacks. It is concluded that the hornblende rims formed contemporaneously with the growth of the large garnets by a process of metamorphic redistribution of hornblende already present in the rock matrix. Each large garnet, as it grew by recrystallization from originally fine-grained, disseminated garnet, displaced matrix plagioclase and hornblende. Of these the plagioclase displayed considerable mobility and was redistributed in the matrix, but the displaced hornblende was less mobile and accumulated as a rim around the large garnet. This conclusion is based upon the nearly constant volumetric ratio of rim to garnet, the essential identity of rim and matrix hornblende, the volumetric equivalence of rim hornblende to calculated displaced hornblende, and the evidence of plagioclase mobility. Conversely, consideration of chemical composition and textural features argue against rim formation either by reaction or by retrograde destruction of the garnet.

PETROGRAPHY AS AN AID IN DATING TERTIARY VOLCANIC ROCKS*

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In connection with stratigraphic studies and mapping of Tertiary rocks in Wyoming during the last 15 years, about 500 rock samples have been collected, either personally or with the cooperation of various colleagues, both from fossil-bearing beds and from beds whose age and stratigraphic relations could be determined by other means. Thin sections were cut from all specimens and heavy mineral separations were made from some.

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From a study of these specimens it is believed that there is a general correlation between ages and types of volcanic rocks throughout all parts of Wyoming where adequate data have been obtained. Although many exceptions can be found for each generalization, the pattern of rock types, when used with caution and with a sufficient number of representative samples, is useful in dating unfossiliferous pyroclastic rocks, and determining the structural history of various areas. It is not known whether this pattern holds true in adjacent regions, but perhaps by calling it to the attention of others, they may broaden or delimit the following "petrographic calendar":

1. Lower Eocene rocks: contain little volcanic material, but where present, it is commonly hornblende andesite with plagioclase, hornblende, and biotite, deeply weathered.
2. Middle Eocene rocks: contain hornblende andesitic and rhyolitic material, fresher and more abundant than in lower Eocene.
3. Upper Eocene rocks: contain pyroxene andesitic material, characteristically green or olive drab.
4. Oligocene rocks: contain hornblende andesitic material, with plagioclase, hornblende, and biotite fresher and with characteristics different from those in middle Eocene; rocks commonly white.
5. Miocene rocks: contain pyroxene andesitic material, less weathered and distinct from upper Eocene; rocks characteristically have variety of bright-colored grains.
6. Pliocene rocks; contain rhyolitic material; data less adequate than for older Tertiary rocks.

BYSTROMITE, MAGNESIUM ANTIMONATE, A NEW MINERAL

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Antimony ores from Antimonio, Sonora, Mexico contain a new mineral corresponding in chemical composition and structure with magnesium antimonate, MgSb_2O_6 , although in the mineral some of the oxygen is replaced by hydroxyl groups and the lattice is deficient in antimony. The mineral is massive, color light blue-gray, streak white, density 5.4 ± 0.1 ; the only associated mineral is quartz. Tetragonal, (ditetragonal dipyramidal), $a = 4.68 \text{ \AA}$, $c = 9.21 \text{ \AA}$, $c/a = 1.968$, space group $P4/mnm$; the structure is of the trirutile type, and the mineral is isomorphous with tapiolite (FeTa_2O_6) and with many artificial antimonates and tantalates. The name bystromite is proposed for the mineral after Anders Bystrom, Swedish crystal chemist, who worked out the structure of synthetic magnesium antimonate in 1941.

FURTHER ACCOUNT OF THE NATURALLY OCCURRING ANTIMONY OXIDES

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A survey of the literature shows that the alteration products of the ores of antimony are in general classified under one of the following names:

Cervantite— Sb_2O_4 or $\text{Sb}_2\text{O}_3 \cdot \text{Sb}_2\text{O}_5$ —1854.

Stibiconite— $\text{H}_2\text{Sb}_2\text{O}_6$ or $\text{Sb}_2\text{O}_4 \cdot \text{H}_2\text{O}$ —1832.

Hydroromeite— $(\text{Sb}^{+++}, \text{Ca}^{++})_2\text{O}(\text{O}_2, (\text{OH})_2) \cdot 2\text{Sb}_2\text{O}_3$ —1932.

The study of over fifty specimens from widely scattered localities indicates that not all of these names are valid. X-ray data show the existence of only one phase, which is sometimes admixed with valentinite, Sb_2O_3 . By spectrographic analysis, this phase has a range in CaO up to 22%. The phase is isometric, the dimension of the unit cell being 10.27 \AA . The index of refraction ranges from 1.66 to $1.9+$, the density ranges from 3.2 to 5.4 and the H_2O content varies from 2% to 13%. When the CaO content is low, admixed valentinite is present, suggesting that calcium is an essential constituent of the phase.

The present work on the homogeneous phase indicates a formula corresponding to $(\text{Ca}, \text{Sb}^{\text{III}})\text{O}-2(\text{Sb}^{\text{V}}2-\text{X})\text{O}, \text{OH}, \text{H}_2\text{O})_{6-7}$ where X varies from 0.-0.5.

THE CRYSTAL CHEMISTRY OF MONTMORILLONITE. II—CALCULATION OF THE STRUCTURAL FORMULA

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As a sequel to an earlier paper describing a new hypothetical structure for montmorillonite, a scheme for calculating the contents of the elementary cell is presented. The method of calculation assumes an integral number of large anions, $(\text{O}+\text{OH})=24$, and relegates the analytically determined cations (including hydrogen) to the basis of 48 charges in order to obtain the unit-cell contents. The assumptions involved in this method of calculation are discussed. The calculations are applied to a new analysis of highly purified montmorillonite for which water determinations are available between 305° and 760° C. and above 760° C. The results are consistent with the hypothesis postulating the occurrence of hydroxyl ions in the tetrahedral layer of this clay mineral.

IMMERSION LIQUIDS OF HIGH REFRACTIVE INDEX

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U. S. Geological Survey, Washington, D. C.

A study of inorganic, organic, and metal-organic compounds that might be employed in the preparation of high refractive index liquids has been undertaken.

Mixtures of arsenic tribromide and precipitated sulfur yielded a series of clear yellow liquids (mixture A), stable at room temperature. An index of 1.829 was obtained. After a period of 12 weeks, the maximum change in index was 0.003. The mixing curve of mixture A with methylene iodide ($n=1.74$ to 1.81) deviated considerably from a straight line.

At room temperature, mixture A and arsenic disulfide formed a series of clear green-yellow to green-amber liquids (B) whose indices range from 1.876 to 1.952. The maximum change in index after a period of 12 weeks was 0.004 for those liquids below 1.92 and 0.007 for those above 1.92. When the temperature was raised during preparation, liquid B was obtained with $n=2.006$. The mixing curve of A and B is a straight line ($n=1.81$ to 2.01).

Red mercuric sulfide added to mixture B gave a clear yellow and very viscous liquid whose index was 2.035

Black selenium added to mixture B gave a clear, light-red semiviscous liquid whose index was 2.017.

Arsenic tribromide, arsenic disulfide, and black selenium formed a very dark red, viscous liquid whose index was 2.008.

All of the liquids described give an acid reaction and decompose some minerals.

GEOLOGY OF THE BRANFORD-KILLINGWORTH AREA, CONNECTICUT†

HARRY M. MIKAMI AND RALPH DIGMAN

E. J. Lavino Co., Norristown, Pennsylvania—State University of New York, Endicott, New York

In the southwestern corner of Connecticut's Eastern Highland section the dominant structural and petrologic features are two stock-like massifs of granite and quartz monzonite and an older and larger pluton of biotite tonalite. The tonalite pluton contains

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gradational concentric changes in fabric from hypidiomorphic-granular to allotriomorphic-gneissic and compositional changes of the plagioclase and hornblende content. The three bodies contain flowage features which describe domal structures. These structures parallel the peripheral contacts, although a zone of mixed rocks is present in the outer part of each intrusive.

The pre-Triassic Middletown and Bolton formations, into which the plutons intruded, are principally metasedimentary rocks. The former is marked by the presence of hornblende and comprises various gneisses formed from interlayered basaltic tuff and pelitic-psammitic sediments. The Bolton formation lies stratigraphically above the Middletown gneiss and consists of schists and quartzites. Contact between the two formations is conformable. Foliation and layering in both are parallel. Both formations contain much pegmatite in bodies ranging from tiny stringers to hundred-foot lenses.

The northwest corner of the area investigated is underlain by sediments and two lava flows of the Triassic Newark group. The eastern border fault, which separates this group from the crystalline rocks, is exposed at one place where a westward dip of approximately 55 degrees was measured. Oblique faults within the Triassic Lowland area and dip abnormalities, including "reverse drag" and half-basin folds in the Newark beds, are discussed.

THE SOLUBILITY OF QUARTZ AND SOME OTHER SUBSTANCES IN SUPERHEATED STEAM AT HIGH PRESSURES

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This paper is a summary of several studies in each of which the solubility of a solid in superheated steam was determined. The results are at 400° and 500° C., and at pressures ranging from 1000 to 30,000 psi. One series of gases are those in equilibrium with liquids at the saturation surfaces in the system $H_2O-Na_2O-SiO_2$. Other results are in two-phase systems gas plus solid, in which the solids include quartz, NaCl, Li_2SO_4 , Na_2SO_4 , K_2SO_4 , $CaSO_4$, Fe_2O_3 and some sulfides.

SYNTHETIC SODIUM-, AMMONIUM-, AND HYDROGEN-URANOSPINITES

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Compounds isostructural with meta-torbernite-I and having the formula $A_2(UO_2)_2(AsO_4)_2 \cdot 8H_2O$ where $A = Na$ or NH_4 , have been synthesized by base exchange with hydrogen-uranospinite, $H_2(UO_2)_2(AsO_4)_2 \cdot 8H_2O$. Hydrogen-uranospinite is ditetragonal-dipyramidal; crystals are flat tablets on {001} with {010} and {110}. X-ray study by the Weissenberg method gave the space group $P4/nmm$ and cell dimensions a_0 7.16 Å, c_0 8.80 ($a_0:c_0=1:1.230$). Specific gravity 3.55 (meas.), 3.55 (calc.). Color lemon-yellow. Perfect cleavage {001}. Optically uniaxial negative with $nO=1.612$ (pale lemon-yellow), $nE=1.584$ (nearly colorless). Sodium-uranospinite has cell dimensions $a_0=7.12$ Å, $c_0=8.70$ ($a_0:c_0=1:1.222$), obtained by powder method. Specific gravity 3.71 (calc.). Optically uniaxial negative with $nO=1.617$ (pale yellow), $nE=1.586$ (colorless). Ammonium-uranospinite has cell dimensions $a_0=7.21$ Å, $c_0=8.85$ ($a_0:c_0=1:1.227$), by powder method. Specific gravity 3.60 (calc.). Optically uniaxial negative with $nO=1.611$ (pale yellow), $nE=1.601$ (colorless). All fluoresce bright lemon-yellow in long-wave ultraviolet radiation and less brightly in short-wave ultraviolet radiation. The relation of these substances to natural troegerite and uranospinite is discussed.

HYDRATION AND BASE EXCHANGE PROPERTIES OF CARNOTITE, TYUYAMUNITE
AND RELATED COMPOUNDS*

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Hydrated uranyl vanadates of potassium, ammonium, thallium, calcium, strontium, barium, sodium, copper, and zinc were synthesized as part of a mineralogic study of carnotite and tyuyamunite. Microscopic and electron-microscopic examination show the crystals to be diamond-shaped plates with micaceous cleavage.

On the basis of x -ray powder patterns, these compounds may be classified into two structural types:

Type I. Compounds of K, NH_4 , and Tl.

Type II. Compounds of Ca, Sr, Ba, Na, Cu, and Zn.

The two types of compounds also differ greatly in the degree of hydration. Type I compounds contain about 1 molecule of water in the air-dried state; type II compounds, 5 to 9 molecules. The higher hydration of type II compounds may be correlated with the presence of low-angle, basal reflections in their x -ray powder patterns.

The water molecules and the characteristic cations (K, NH_4 , Ca, etc.) occur between uranyl vanadate layers, a condition analogous to that found in the clay mineral montmorillonite. There is a progressive decrease in the interlayer spacing of type II compounds as water is removed by heating.

Type II compounds, when treated with solution of potassium salts, rapidly exchange their cations for potassium and are converted to carnotite. This entails a contraction of the interlayer spacing as the lower hydration characteristic of type I is assumed. The reverse process, the conversion of carnotite to compounds of type II, involves the expansion of interlayer spacing, and takes place much less readily.

A SECOND SOUTH AMERICAN OCCURRENCE OF BRAZILIANITE

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In the pegmatite of the *alto* Patrimonio, at Piedras Lavradas, Paraíba, Brazil, massive amblygonite was found to be cut by veins and patches of brazilianite accompanied by wardite, apatite, and other phosphates. The brazilianite occurs in small (1–2 mm.) grains and crystals, some fairly well developed. Measurable crystals show the following forms: {110}, {010}, {320}, {130}, {670}?, {101}, {301}, {021}? {111}, {321}, {121}. The mineral was identified by indices, crystal form, and x -ray pattern. The associated wardite is somewhat coarser grained, shows a good cleavage, and is sometimes in crude crystals. In the veins, wardite is earliest, followed by brazilianite (usually idiomorphic) and purplish apatite. In the patches, a very finely fibrous greenish mineral, resembling hydroxy-apatite; wardite, brazilianite and apatite follow in that order, with some overlapping, and all are covered by a very fine grained compact white mineral which may be meta-variscite.

X-RAY EXAMINATION OF URANOTHORITE

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Earlier x -ray studies of thorite have indicated that it is metamict (Vegard, *Phil. Mag.*, 32: 65, 1916) or have led to cell dimensions (Boldyrev et al., Leningrad, *Inst. Mines, Ann.*, 11: 1, 1938) that seem very much out of line with those of related materials.

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Single crystal *x*-ray diffraction patterns have been obtained from unaltered uranothorite from Gillespie's Beach, South Westland, New Zealand. Rotation patterns on the *a* and *c* axes are very similar to zircon patterns but show a certain amount of streaking of the spots along "powder arcs." A cell of dimensions $a_0 = 7.12 \text{ \AA}$, $c_0 = 6.32 \text{ \AA}$ contains 4ThSiO_4 . Th is substituted by U in the ratio of about 1/7. Observed and calculated densities agree at 6.7 ± 0.05 . The space group is $D_{4h}^{19} - I4/amd$. The similarity of the rotation patterns to those of zircon leaves no doubt that the structure of this uranothorite is of the H3 (zircon) type.

ANION EXCHANGE IN CLAY MINERALS

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The amount and nature of the anion exchange in various clay minerals were determined using electrodialyzed materials. The effect of the chloride, sulphate, and phosphate ions was determined and compared with the base exchange capacity of these clay minerals. A tentative explanation regarding the ion exchange phenomena was developed as a result of this study. It is suggested that in kaolinite and illite the ion exchange, both anionic and cationic, takes place on "active spots" primarily due to broken valence bonds. In the case of montmorillonite, where cation exchange may result also from excess negative lattice charge, it is suggested that a similar exchange may take place with anions due to excess positive charge. As a consequence of this, clays probably are not amphoteric in the usual chemical sense.

As further evidence, additional data were obtained on the effect of particle size on the cation and anion exchange capacity of these clay minerals. The anion exchange capacity in kaolinite was shown to depend on the lateral surface area of the clay mineral. The relationship existing between anion exchange capacity and lateral surface area was also demonstrated for other clay minerals.

FURNACE ATMOSPHERE CONTROL IN DIFFERENTIAL THERMAL ANALYSIS*

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The usefulness of differential thermal analysis can be extended considerably by controlling the composition of the furnace atmosphere. The method described for securing atmosphere control is applicable to most existing differential thermal analysis furnaces. Examples are given which show the effect of a furnace atmosphere of nitrogen on differential thermal curves of clays containing organic matter and pyrite. Other curves show the effect of CO_2 on differential thermal curves of siderite, magnesite, dolomite and calcite. One curve follows the alternate dissociation-reconstitution of the CaCO_3 part of dolomite in an atmosphere of CO_2 . The effect, on the differential thermal analysis curve, of filling the furnace with a gas which is a participant in the reaction is explained by the relation of the partial pressure to the equilibrium constant and the heat of reaction.

ORIGIN OF A LAYERED ULTRAMYLONITE FROM SOUTHEASTERN CONNECTICUT

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At the southernmost contact of the Preston gabbro is a zone of imbricate structure which the hydrothermally altered gabbro and four tabular bodies of chlorite-actinolite,

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schist derived from it have been brought into juxtaposition with a block of medium-rank gneiss and its progressively mylonitized equivalents by reverse faulting. The lower five feet of the block of non-layered biotite-cummingtonite-hornblende gneiss was converted to a layered slate-like ultramylonite composed of alternating quartz-plagioclase laminae and biotite-amphibole laminae which range in thickness from a thin film to five millimeters. The ultra mylonite has a well-developed cleavage parallel to the lamination. The temperature which prevailed during mylonitization was apparently high enough to prevent significant diaphoresis and low enough to prevent recrystallization. The block is at least twenty feet thick and it exhibits all stages of cataclastic reduction and increasing metamorphic differentiation from top to bottom. No transposition of s-planes was involved and the dip of the foliation in all the cataclastic rocks is parallel to the uniformly dipping reverse faults. The ultramylonite was subjected to post-layering microfaulting.

The origin of the layering is discussed in the light of the solution principle of Eskola, the mechanical hypothesis of Schmidt and Wenk, and the complex hypothesis of Sander and Turner. It is concluded that the process of metamorphic differentiation was dominantly mechanical and a consequence of the difference in mechanical behavior of the light and dark minerals under shearing stress. The degree of perfection of the lamination is believed to be a function of the kinematics of the mass during mylonitization.

RELATION OF TYPE OF COUNTRY ROCK TO THE SHAPE OF GRANITIC PEGMATITE INTRUSIONS*

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Examination of more than 1800 pegmatite intrusions in the Quartz Creek district, Gunnison County, Colorado, has shown that the shape of a granite pegmatite intrusion is controlled by: (1) the type and competency of country rock; (2) the composition of the pegmatite magma; and (3) the amount of intruded material. Of these, the first is commonly the most important.

Intrusions of pegmatite in competent rocks, such as granite, monzonite, quartzite, and hornblende gneiss, commonly cut across the older layered structures such as bedding, banding, or foliation, and follow joints or other fractures, forming thin, tabular to sinuous, branching, and irregular bodies.

The pegmatites in incompetent rocks, such as mica schist, are in general concordant with foliation and were intruded by shouldering apart the country rock along foliation planes. Pegmatites of this type are commonly lenticular, troughlike, arcuate, or tear-drop-shaped; branching and irregular shapes are rare.

The pegmatites in the competent rocks tend to be thinner than those in the incompetent rocks; thickly lenticular pegmatites are practically unknown.

All bodies of pegmatite with the same composition do not have the same shape, although in some districts lithium-bearing pegmatites tend to be bulbous. With the intrusion of large amounts of pegmatitic material, the effect of the country rock on the shape of the pegmatite is usually obscured and the body assumes an irregular stock-like shape. The type of country rock thus appears to have the greatest control over the final shape of the smaller pegmatite bodies.

Similar relations between the shape of pegmatites and the type of country rock have been found in other districts.

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OBSERVATIONS ON TWIN LAWS COMMONLY EXHIBITED BY PLAGIOCLASE OF METAMORPHIC ROCKS

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Observations on the twin laws commonly exhibited by metamorphic (especially by deformed) rocks are placed on record. Attention is drawn to the marked contrast between the complex association of twins typical of plagioclase in igneous rocks, and the much less complex twinning of metamorphic plagioclase, as a possible basis for distinguishing (1) metasomatic from magmatic plagioclase in granitic rocks; (2) relict from metamorphic plagioclase in amphibolites.

GROWTH OF WATER SOLUBLE AND OTHER CRYSTALS AT LOW TEMPERATURES

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Crystals have been synthesized at low temperatures from (1) water solutions, (2) melts with or without temperature gradient conditions, and (3) hydrothermal processes. Extensive investigations have been made in crystal synthesis directly from aqueous solutions. Outstanding success has been attained with Rochelle Salts, ammonium dihydrogen phosphate and ethylene diamine tartrate, by various investigators in the United States and elsewhere. Numerous other inorganic and organic crystals have been grown readily. Although few of these crystals occur in nature, the principles involved in their synthesis are of interest to the mineralogist and geologist because the mechanics of growth may be similar to those of natural water soluble crystals. It is of more than passing interest however, that NaCl crystals of large size are crystallized commercially from melt at high temperatures. Certain organic crystals have been synthesized by crystallization from a low temperature melt using a temperature gradient method similar to that employed successfully for commercial synthesis of NaCl and CaF_2 at higher temperatures. The Signal Corps Engineering Laboratories at Fort Monmouth, New Jersey, have been engaged actively in synthesis of berlinite (AlPO_4) since 1946. More recently this organization has synthesized AlAsO_4 . Both of these crystalline materials are synthesized hydrothermally at or below 250°C .; both are unusual in that they have retrograde solubility characteristics. Tourmaline has been synthesized on a small scale from natural source materials at low temperatures for the Signal Corps by Baird Associates, Cambridge, Massachusetts. Considerable data have been accumulated showing that quartz crystal can be synthesized at low temperatures (250°C .). Although synthesis of the last two minerals at low temperatures has no practical value at this time, geologic implications are evident.

HUMMERITE AND MONTROSEITE, TWO VANADIUM MINERALS FROM MONTROSE COUNTY, COLORADO*

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Two new minerals, collected in the summer of 1949, are described and compared with previously known vanadium minerals from Colorado.

Hummerite was first identified in bright-orange veins in gray clay at the Hummer mine of the Jo Dandy Group on the southwest side of Paradox Valley, Montrose County, Colorado, and later as a yellow efflorescence on sandstone at the North Star mine on the

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opposite side of Paradox Valley. It is translucent, water soluble, and similar in appearance to pascoite but has the composition hydrous magnesium vanadate. The x -ray powder pattern of crystals formed by evaporation of a water solution is the same as that of the original vein mineral. The recrystallized material is triclinic, pinacoidal class, and forms crystals elongated parallel to the c axis or tabular parallel to the b and c axes. It is optically negative, has strong dispersion, and $n_Y = 1.81$. The chemical analysis and morphologic and x -ray crystal data will be presented.

Montroseite, in black microscopic bladed crystals, was found in the Bitter Creek mine on the northeast side of Paradox Valley, Montrose County, Colorado. It is orthorhombic, probably rhombic-dipyramidal class, opaque, and has the composition $2\text{FeO} \cdot \text{V}_2\text{O}_5 \cdot 7\text{V}_2\text{O}_4 \cdot 4\text{H}_2\text{O}$. Morphologic and x -ray crystal data are presented together with spectrographic and chemical analyses.

SURFACE STRUCTURE AND CRYSTAL GROWTH

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The concept, that the cube face of a crystal having NaCl structure consists of a checkered pattern emanating positive and negative force fields, represents an idealization which is useless for explaining the surface properties of a large number of crystals. It is not understandable on this basis why sintering of calcined MgO should require the very high temperature it does and why it can be accomplished at a much lower temperature if certain "impurities" are present. An explanation for this and related phenomena is presented on the basis that crystals containing ions of widely different polarizabilities develop surfaces which contain primarily the most polarizable ion in the extreme outer layer, followed by the less polarizable ion which is slightly recessed. This deviation of the surface from the bulk structure can lead to an electrical double layer setting up repulsion forces between the single particles and thus preventing sintering.

The surface structure depends largely on the surrounding medium, a fact which can be demonstrated by the change in the hydrophilic and the catalytical properties of PbF_2 and HgI_2 on removal of the water layer.

These concepts are applied to the crystallization of substances from various solvents as affected by impurities.

PETROLOGY OF THE ALKALINE ROCKS OF THE NEMURO DISTRICT, HOKKAIDO, JAPAN

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In the Nemuro Peninsula, alkaline rocks occur in the Cretaceous formations as sheets, laccoliths, or rarely as submarine flows. Their age is probably upper Cretaceous or lower Tertiary. The rocks vary from picritic trachydolerite, trachydolerite, and trachybasalt, to monzonitic and syenitic rocks. Minerals present, which often show zonal structures, include plagioclase, anorthoclase, orthoclase, analcite, natrolite, thomsonite, augite, aegirine-augite, olivine, biotite, and iron ores. The preponderance of K_2O over Na_2O is remarkable, and the Peacock alkali-lime index is 52.1.

The margins of thick sheets or laccoliths are chilled porphyritic trachybasalt. Dark picritic trachydolerite rich in olivine and augite lies immediately above the lower chilled margin and grades upward into less dark trachydolerite and monzonitic rocks near the upper chilled margin. The materials only when the chilled margins are occasionally cut by syenitic veins varying in thickness from a few inches to a foot. Thus gravitational differentiation *in situ* is inferred to have occurred in these intrusive bodies.

Various parts of the pillow structure, which is sometimes well developed in the thinner sheets, show noticeable differences in texture as well as in chemical composition. It is argued that crystallization in an extremely water-rich magma rather than under a water cover is responsible for the formation of the pillows.

The original magma of these alkaline rocks was probably shonkinitic in composition, with abundant volatile components, which played an important role in crystallization, especially in the late magmatic and hydrothermal stages.

COMPLETE SUBSTITUTION OF ALUMINUM FOR SILICON: THE SYSTEM



H. S. YODER AND M. L. KEITH

Geophysical Laboratory, Washington, D. C.

Thermal, optical, and x-ray data indicate that there is a complete series of solid solutions between spessartite, $\text{Mn}_3\text{Al}_2(\text{SiO}_4)_3$ and yttrogarnet, $\text{Y}_3\text{Al}_2(\text{AlO}_4)_3$. The substitution $\text{Y}^{+3}\text{Al}^{+3} \rightarrow \text{Mn}^{+2}\text{Si}^{+4}$ may be represented by the formula:



Yttrogarnet has a cube edge of 12.01 ± 0.02 Å and the space group O_h^{10} . Using the coordinates of grossularite as determined by Menzer, good agreement was obtained between calculated and observed intensities. The atomic positions are:

Al in 16(a)	0	0	0
Y in 24(c)	$\frac{1}{4}$	$\frac{1}{8}$	0
Al in 24(d)	$\frac{1}{4}$	$\frac{3}{8}$	0
O in 96(h)	x	y	z

where x, y, and z are 0.04, 0.055, and 0.64 respectively. The yttrium is in eight-fold coordination; the aluminum is in both four- and six-fold coordination. Yttrogarnet inverts to a high form at approximately $1970 \pm 50^\circ$ C.

This is the first case on record of the complete substitution of aluminum for silicon in an orthosilicate. It contradicts the prevalent idea that the substitution of aluminum for silicon always decreases as the Si/O ratio decreases.

THE MECHANISM OF VEIL FORMATION IN CRYSTALS*

S. ZERFOSS

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Numerous observers have reported that there is a maximum practical rate of crystal growth. Crystals grown at faster rates invariably develop flaws—veils. These veils are lines, planes or bodies of liquid and gaseous inclusions commonly oriented with respect to some growing face. One expects, that when a seed is planted in a saturated solution, there will be considerable veiling to make the adjustment of the seed to the equilibrium endform of the crystal. Thereafter when the endform is in equilibrium with the solution, growth at a slow rate should proceed with a high degree of perfection. Experience with laboratory growth of numerous crystals together with pilot plant growth of ADP is drawn upon to elaborate a mechanism of veil formation and of crystal growth from water solution. Examples of natural crystals can be used to support the general picture.

* The material in this talk will be published under the authorship of P. H. Egli, S. Zerfoss and S. Slawson.

LIST OF FORMER OFFICERS AND MEETING PLACES

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| 1920 Chicago, Illinois | 1936 Cincinnati, Ohio |
| 1921 Amherst, Massachusetts | 1937 Washington, D. C. |
| 1922 Ann Arbor, Michigan | 1938 New York, N. Y. |
| 1923 Washington, D. C. | 1939 Minneapolis, Minnesota |
| 1924 Ithaca, New York | 1940 Austin, Texas |
| 1925 New Haven, Connecticut | 1941 Boston, Massachusetts |
| 1926 Madison, Wisconsin | 1942 No meeting held |
| 1927 Cleveland, Ohio | 1943 No meeting held |
| 1928 New York, N. Y. | 1944 No meeting held |
| 1929 Washington, D. C. | 1945 Pittsburgh, Pennsylvania |
| 1930 Toronto, Canada | 1946 Chicago, Illinois |
| 1931 Tulsa, Oklahoma | 1947 Ottawa, Canada |
| 1932 Cambridge, Massachusetts | 1948 New York, N. Y. |
| 1933 Chicago, Illinois | 1949 El Paso, Texas |
| 1934 Rochester, New York | 1950 Washington, D. C. |
| 1935 New York, N. Y. | |

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MINERALOGICAL SOCIETY (LONDON)

The anniversary meeting of the Society was held on Thursday, November 2nd, 1950, in the apartments of the Geological Society of London, Burlington House, Piccadilly, W.1 (by kind permission).

The following papers were read:

(1) PRIDERITE, A NEW MINERAL FROM THE LEUCITE LAMPROITES OF THE WEST KIMBERLEY AREA, WESTERN AUSTRALIA.

By Mr. K. Norrish (communicated by Prof. R. T. Prider).

Priderite is the name suggested for a new mineral with the formula $(K, Ba)_{1.3}(Ti, Fe)_8O_{16}$. X-ray diffraction studies show that the mineral is body centered, tetragonal with $a = 10.11$ and $c = 2.96\text{\AA}$. Structurally the mineral resembles cryptomelane KMn_3O_{16} , the Ti of priderite being equivalent to the Mn of cryptomelane. Synthetic K- and Ba-priderites appear to be isostructural and they are probably members of a series analogous to cryptomelane-hollandite.

(2) SOME NOTABLE HABIT CHANGES IN BARYTES FROM WESTMORLAND AND DURHAM.

By Mr. W. F. Davidson and Mr. A. F. Seager.

A number of barytes specimens recently discovered in the Silver Band mine, Westmorland, show marked habit changes, associated with beautiful color zoning. Initially, the crystals were elongated on $[a]$ with a large development of $\{011\}$ and $\{001\}$. The final habit is tabular on $\{001\}$, with $\{110\}$ and $\{102\}$ as subsidiary forms. Some specimens show fluorescence. A specimen from the Cow Green mine, Teesdale, Co. Durham, is also described.

(3) NEW OCCURRENCES OF DUFTITE.

By Dr. G. F. Claringbull.

During an examination of bayldonite and other copper lead arsenates, a specimen of supposed bayldonite from Mapimi, Mexico, was found to be isomorphous with descloisite and to give an x-ray powder pattern near to but not identical with conichalcite. Other specimens from localities in Cumberland sent for identification by Mr. W. F. Davidson give patterns matching that of the Mapimi material and all are closely similar to duftite $(CuPbAsO_4OH)$ from Tsumeb, S.W. Africa. Chemical tests confirm the presence of Cu and Pb.

(4) THE BREECE, NEW MEXICO, METEORITIC IRON.

By Dr. Carl W. Beck, Dr. Lincoln LaPaz and Mr. Louis H. Goldsmith.

The investigation of this siderite (50 kg., found 1921) shows it to be a medium octahedrite composed chiefly of kamacite with lesser amounts of taenite and plessite fields. The most interesting constituent is cohenite as well defined lamellae in the unusual arrangement similar to the familiar Reichenbach and Brezina lamellae.

The following were taken as read or read in abstract:

(5) JACOBSITE FROM THE TAMWORTH DISTRICT OF NEW SOUTH WALES.

By Dr. F. L. Stillwell and Mr. A. B. Edwards.

(6) THE USE OF THE GNOMONIC PROJECTION IN THE DETERMINATION OF THE INDICATRIX OF CRYSTALS.

By Mr. N. Joel.

(Titles and abstracts kindly submitted by G. F. Claringbull, General Secretary.)

NOTES AND NEWS

NOTES ON SOME CALIFORNIA MINERALS

NUEVITE=SAMARSKITE; TRONA AND HANKSITE; GAYLUSSITE

JOSEPH MURDOCH, *University of California at Los Angeles.*

Nuevite = Samarskite

In 1946 the writer reported a samarskite-like mineral from the Southern Pacific Silica Quarry near Nuevo, Riverside County, California, as a new species, giving it the name "nuevite" (1). This determination was made on the basis of a spectroscopic analysis showing no uranium and a small amount of tantalum, and rough crystals which gave an apparent axial ratio differing from samarskite. The general physical and optical properties put the mineral in the samarskite group but the apparent absence of U and Ta seemed to warrant a new species name. However, after considerable delay, it was possible to have a chemical analysis made, which at once showed the material to be samarskite, a finding confirmed by Dr. Herman Yagoda (private communication), who kindly made a polished surface and showed uranium comparable to a U-rich variety of samarskite. Accordingly, the name nuevite should be considered as invalidated, and the occurrence recorded as samarskite. The roughly formed crystals, in revised orientation, showed measurements in reasonable agreement with samarskite.

As is usually the case, it is metamict, and recently the writer had an opportunity to take an x-ray powder photograph of the ignited material, which showed a typical samarskite pattern, according to Dr. Clifford Frondel (private communication). It has seemed worthwhile to record the chemical analysis, and the information given by this powder photograph, since none has apparently been published up to the present time. The x-ray powder data represent figures corrected for camera radius and film shrinkage, in Å units.

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I wish to express my thanks to the following for assistance and advice during this determination: Miss Jewel J. Glass, U. S. Geological Survey; Professor Clifford Frondel, Harvard University; Dr. Herman Yagoda, U. S. Public Health Service; Professor George Tunell, University of California at Los Angeles. The chemical analysis was made possible by a grant from the research funds of the University of California.

Trona and Hanksite

Recently a fragment of crystalline salines was collected from a dried pool on the east side of Mono Lake, California, and given to the writer

TABLE 1. X-RAY POWDER SPACING DATA FOR SAMARSKITE, NUEVO, CALIFORNIA
COPPER RADIATION, NICKEL FILTER, SPACINGS IN Å UNITS

<i>I</i>	<i>d/n</i>	<i>I</i>	<i>d/n</i>
2	10.97	1	1.65
$\frac{1}{2}$	5.61	1	1.59
3	3.68	2	1.537
1	3.53	$\frac{1}{2}$	1.519
$\frac{1}{2}$	3.40	4	1.502
1	3.25	1	1.436
$\frac{1}{2}$	3.13	1	1.390
10	3.07	$\frac{1}{2}$	1.355
10	2.92	$\frac{1}{2}$	1.301
2	2.80	$\frac{1}{2}$	1.279
$\frac{1}{2}$	2.71	$\frac{1}{2}$	1.229
4	2.59	$\frac{1}{2}$	1.218
2	2.47	$\frac{1}{2}$	1.193
2	2.44	$\frac{1}{2}$	1.177
$\frac{1}{2}$	2.23	$\frac{1}{2}$	1.127
1	2.17	$\frac{1}{2}$	1.117
$\frac{1}{2}$	2.12	$\frac{1}{2}$	1.091
1	2.08	$\frac{1}{2}$	1.081
3	1.90	$\frac{1}{2}$	1.047
4	1.84	$\frac{1}{2}$	1.008
1	1.76	$\frac{1}{2}$.9823
1	1.75	$\frac{1}{2}$.8995
1	1.73	$\frac{1}{2}$.8633
1	1.71	$\frac{1}{2}$.8202
$\frac{1}{2}$	1.67		

TABLE 2. ANALYSIS OF SAMARSKITE, NUEVO, CALIFORNIA

SiO ₂	.28
Ta ₂ O ₅	22.08
Cb ₂ O ₅	32.46
TiO ₂	2.75
Y ₂ O ₃	11.98
Ce ₂ O ₃	tr.
UO ₂	13.66
UO ₃	3.51
FeO	11.09
MnO	.57
ZrO ₂	.02
PbO	.14
Loss on ignition in N	1.07
	<hr/>
	99.61

Analysis by W. H. Herdsman

by his colleague, Professor W. C. Putnam. The specimen is roughly stratified, with one layer massive or spongy halite, and the other a faintly pinkish aggregate of flattened and elongated crystals, in part covered with a white finely crystalline and sometimes spherulitic coating. The larger crystals, ranging in length up to one centimeter, were found to be trona, which has not been previously reported from Mono Lake. The finely crystalline coating turned out to be hanksite, hitherto found only at Searles Lake. The sequence of deposition is halite, then trona, then hanksite.

The trona, even in perfectly fresh clear crystals, showed a strong potassium flame when the sodium flame was cut out with a Merwin screen. The crystals are elongated parallel to the b axis and usually somewhat irregularly flattened parallel to the base. They are invariably terminated by a single form, $\{\bar{1}11\}$, but show a great variety of faces in the orthodome zone. Of this series, $\{100\}$ is usually of good quality and fair size. The other faces are present often as narrow or line faces in a zone, or as curved surfaces, but sometimes well developed. A list of forms observed in good position and of reasonable quality is as follows: $\{\bar{1}11\}$, $\{100\}$, $\{001\}$, $\{\bar{7}01\}$, $\{\bar{1}02\}$, $\{\bar{1}03\}$, $\{\bar{1}04\}$, $\{\bar{5}01\}$, $\{302\}$, $\{401\}$. Other possible forms, as line faces only, or a little off position include $\{105\}$, $\{106\}$, $\{\bar{1}05\}$, $\{\bar{1}09\}$, $\{\bar{1}.0.10\}$, $\{1.0.12\}$, $\{\bar{1}.0.15\}$.

The hanksite in the specimen is not in recognizable crystals, but was identified by its uniaxial negative character, index of refraction, $\omega = 1.48 \pm$, strong birefringence, and parallel extinction. Chemical tests showed effervescence with acid and precipitates of BaSO_4 with BaCl_2 and AgCl with AgNO_3 . Flame coloration showed strong potassium reaction.

Gaylussite

Published data on the crystallography of gaylussite depend on the measurements of a single crystal from Venezuela, made by W. Phillips (2) in 1827. Crystals described from Searles Lake, California, by J. H. Pratt (3) in 1896, were not suitable for measurement on the reflecting goniometer, but the angles were checked on larger crystals with a contact goniometer. Through the courtesy of Mr. L. J. Bailey, chemist at the American Potash and Chemical Company, Trona, California, the writer has been enabled to study some excellent crystals of gaylussite from Searles Lake, and to offer some refinements in the values of the crystallographic elements.

The crystals were very suitable for measurement on the two-circle goniometer, and the results on five selected crystals furnished values quite close to those of Phillips, thus confirming the quality of his work.

There are, however, small but consistent variations from his values, and since the present figures represent the average of a number of good readings as compared with a single set it was considered worth while to recalculate the crystallographic elements, and present a new angle table.

The Searles Lake crystals show the characteristic habit for this occurrence, with $\{001\}$ and $\{\bar{1}12\}$ dominant, $\{110\}$, $\{001\}$, and $\{\bar{1}01\}$ smaller, and $\{010\}$ appearing as a poor face on only one crystal. The averages of best readings for each face are listed in Table 1, with Phillips' values for comparison. Calculated values are given in the revised angle table (Table 2).

TABLE 1. MEASURED ANGLES FOR GAYLUSSITE

Quality of signal	$\{001\}$		Quality of signal	$\{\bar{1}01\}$	
	ϕ	ρ		ϕ	ρ
<i>B</i>	89°42'	12°08'	<i>C</i>	-90°00'	37°58'
<i>B</i>	90 07	12 14	<i>B</i>	-90 09	38 14
<i>B</i>	89 39	11 52	<i>B</i>	-89 59	37 48
<i>C</i>	90 35	11 48			
				av. new	38 00
	av. new	12 00½		old	38 08
	old	11 33			
	$\{110\}$			$\{\bar{1}12\}$	
	ϕ	ρ		ϕ	ρ
<i>B</i>	34°18'	90°00'	<i>C</i>	-21°29'	37°56'
<i>B</i>	34 20	90 00	<i>B</i>	-21 30	37 56
<i>B</i>	34 23	90 00	<i>C</i>	-21 35	37 50
<i>B</i>	34 43	90 00	<i>D</i>	-21 30	37 50
<i>B</i>	34 35	90 00	<i>C</i>	-21 08	37 51
<i>B</i>	34 34	90 00	<i>D</i>	-21 04	37 51
av. new	34 29		av. new	-21 22½	37 52
old	34 25		old	-21 55	37 53
	$\{011\}$				
	ϕ	ρ			
<i>B</i>	8°11'	55°45'			
<i>C</i>	8 25	55 38			
<i>C</i>	8 12	55 32			
<i>C</i>	8 25	55 19			
<i>C</i>	8 17	55 30			
<i>C</i>	8 32	55 34			
<i>C</i>	8 11	55 31			
av. new	8 19	55 33			
old	8 03	55 34			

TABLE 2. ANGLE TABLE FOR GAYLUSSITE

Monoclinic	$a:b:c=1.4878:1:1.4453$			$\beta = 102^{\circ}00\frac{1}{2}'$		
	$p_0q_0:1=0.97413:1.4137:1$			$\mu = 77^{\circ}59\frac{1}{2}'$		
	$r_2:p_2:1=0.70737:0.68716:1$					
	$p_0'=0.99316, q_0'=1.4453$			$x_0'=0.21725$		
Form	ϕ	ρ	ϕ_2	$\rho_2=B$	C	A
$c \{001\}$	$90^{\circ}00'$	$12^{\circ}00\frac{1}{2}'$	$77^{\circ}59\frac{1}{2}'$	$90^{\circ}00'$	$0^{\circ}00'$	$77^{\circ}59\frac{1}{2}'$
$b \{010\}$	$0 \ 00$	$90 \ 00$	—	$0 \ 00$	$90 \ 00$	$90 \ 00$
$a \{100\}$	$90 \ 00$	$90 \ 00$	$0 \ 00$	$90 \ 00$	$77 \ 59\frac{1}{2}$	$0 \ 00$
$m \{110\}$	$34 \ 30$	$90 \ 00$	$0 \ 00$	$34 \ 30$	$83 \ 14$	$55 \ 30$
$e \{011\}$	$8 \ 22\frac{1}{2}$	$55 \ 36\frac{1}{2}$	$77 \ 59\frac{1}{2}$	$35 \ 16\frac{1}{2}$	$54 \ 43\frac{1}{2}$	$84 \ 06$
$s \{\bar{1}01\}$	$-90 \ 00$	$38 \ 01\frac{1}{2}$	$128 \ 01\frac{1}{2}$	$90 \ 00$	$50 \ 02$	$128 \ 01\frac{1}{2}$
$r \{\bar{1}12\}$	$-21 \ 30$	$37 \ 50$	$105 \ 53\frac{1}{2}$	$55 \ 12$	$43 \ 28\frac{1}{2}$	$102 \ 59\frac{1}{2}$

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ON THE MINERALOGY OF ANTARCTICA

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Since 1895 ninety-six papers have been published relating to Antarctic mineralogy and petrography. Dr. Johannes Petersen (1895: 275-278) was the first to publish on Antarctic petrography. He described the basalt of Mount Christen Christensen (Christensen-Vulkan) of Robertson Island, West Antarctica.

The following is a list of the 167 mineral species, subspecies, and varieties, as well as those of questionable occurrence, that have been reported from Antarctica. A number of these have been determined only microscopically, and in the case of gold its presence has been determined only by chemical analysis.

Acmite	Andalusite	Aphrosiderite	Beryl
Actinolite	Andesine	Apophyllite	Biotite
Adularia	Andradite	Arfvedsonite	Bornite
Aegirine-augite	Anomite	Arsenopyrite	Bronzite
Allanite	Anorthite	Atacamite	Brookite
Almandite	Anorthoclase	Augite	Brucite
Analbite?	Anthophyllite	Azurite	Brushite?
Analcite	Antigorite	Barkevikite	Bytownite
Anatase	Apatite	Basaltic hornblende	Calcite

Cancrinite	Forsterite	Mesolite	Scapolite
Cassiterite	Galena	Microcline	Scolecite
Ceylonite	Glauconite	Microperthite	Sericite
Chalcopyrite	Glaucophanes	Mirabilite	Serpentine
Chlor-apatite	Gold	Mizzonite	Siderite
Chloritoid	Graphite	Molybdenite	Sillimanite
Chondrodite	Grossularite	Muscovite	Sodalite
Christensenite	Gypsum	Natrolite	Soda-orthoclase
Chrome diopside	Hedenbergite	Nephelite	Spessartite?
Chromite	Hematite	Newberyite?	Sphalerite
Chrysolite	Hercynite	Nosean	Sphene
Chrysotile	Heulandite	Oligoclase	Stercorite
Clinochlore	Hornblende	Olivine	Stibnite
Clinoenstatite	Hypersthene	Orthoclase	Stilbite
Clinohumite?	Iddingsite	Paragonite?	Talc
Cordierite	Ilmenite	Pargasite	Tetrahedrite
Corundum	Kaersutite?	Penninite	Thomsonite
Cossyrite	Kaolinite	Phlogopite	Titaniferous aegirine-augite
Cristobalite?	Katophorite?	Picotite	
Cumingtonite	Kornerupine?	Piedmontite	Titaniferous augite
Damourite	Kyanite	Pigeonite	Titanomagnetite
Delessite	Labradorite	Pinite	Topaz
Diallage	Laumontite	Pistacite	Tourmaline
Diopside	Lawsonite	Prehnite	Tremolite
Dolomite	Lepidolite	Prochlorite	Tridymite
Dumortierite	Lepidomelane	Pyrite	Vermiculite?
Edenite	Leucite	Pyrrhotite	Vesuvianite
Enstatite	Leucoxene	Quartz	Woehlerite
Enstatite-augite	Limonite	Riebeckite	Wollastonite
Fassaite?	Magnetite	Rutile	Xenotime
Fayalite	Malachite	Salite	Zircon
Ferrimolybdenite	Malacolite	Sanidine	Zoisite
Fluorite	Meionite	Sapphire	

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DISCUSSION OF "X-RAY DIFFRACTION PATTERNS OF ASBESTOS"

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In a recent paper, "X-ray Diffraction Patterns of Asbestos," *Am. Mineral.*, **35**, 579-589 (1950), Suzanne van Dijke Beatty compared the x-ray diffraction patterns of several different types of asbestiform minerals with the purpose of providing data for the identification of these minerals by means of x-rays. Misinterpretations of the data were made because four of the samples studied were improperly identified. Since, in our opinion, the correction which appeared on page 1090 of the November-December 1950 issue is inadequate, we herewith review in greater detail the results reported.

According to our interpretation of Dr. Beatty's data, the four minerals referred to as chrysotile in Tables 6, 7, 8, and 9, are actually amphiboles of the tremolite type. These are the samples which are labeled as being from (1) Corsica, (2) Easton, Pa., (3) Labrador, and (4) Chester, Pa. The d values and measured intensities reported for these samples by Dr. Beatty agree very well with the data given by Johansson for grammatite, a comparatively pure tremolite.¹ To show how the d values remain virtually unchanged by isomorphic replacements in the tremolite structure, the data obtained by x-ray analysis of a synthetic fluor-amphibole containing 12 per cent Al_2O_3 is also reported. These data are compared in Table 1 with the results given by Beatty.

Dr. Beatty correctly grouped together the diffraction patterns of the misidentified samples and noted differences between them and the patterns obtained from the true chrysotiles. Since Dr. Beatty observed that the d values of the eight chrysotile-labeled samples could be arranged into two distinct groups, it is rather surprising that chemical analyses or petrographic examination of samples from each of these groups were not given. It is to be expected that such studies would have shown clearly the marked compositional and optical differences which exist. When x-ray data are presented which are to be used as a reference, it is highly desirable, and usually essential, to report the degree of homogeneity and the optical constants of the material. When this knowledge is used in conjunction with the chemical analysis the significance of every line in the pattern may be better understood.

When the 4 amphibole samples are discounted, the interplanar spacings of chrysotile do not show the considerable variations claimed. The two

¹ Johansson, K., Vergleichende Untersuchungen an Anthophyllit, Grammatit, und Cumingtonit: *Zeit. Krist.*, **73A**, 31-51 (1930).

TABLE 1. COMPARISON OF X-RAY DIFFRACTION DATA OF NATURAL AND SYNTHETIC TREMOLITE WITH MISIDENTIFIED "CHRYSTOLITE"

Labrador ¹		Natural Tremolite (Grammatite ²)		Synthetic Fluorine- Containing Amphibole ³		Miller Indices
<i>d</i> Å. U.	Meas. Int.	<i>d</i> Å. U.	Meas. Int.	<i>d</i> Å. U.	Meas. Int.	
9.0	6			9.0	7	020
8.4	19	8.41	3	8.4	29	110
4.51	4	4.50	2	4.48	8	040
3.37	8	3.38	2	3.38	9	220
3.27	10	3.27	2	3.25	16	240
3.12	32	3.13	4	3.10	43	310
2.94	9	2.94	2	2.94	9	221
2.80	4	2.80	2	2.78	10	330
2.71	16	2.71	4	2.70	16	151
2.67	7					
2.60	6	2.59	2	2.59	8	061
2.54	8	2.52	3	2.53	7	202
2.34	10	2.33	2	2.32	7	351
2.28	4	2.26	1	2.254	2	312
2.169	9	2.16	2	2.161	8	261
2.043	5	2.04	1	2.058	2	202
2.017	8	2.01	2	2.015	5	402
1.897	5					
1.653	8	1.65	2	1.645	9	461
1.585	5½	1.573	2	1.580	3	153
1.514	4	1.506	2			
1.505	4	1.503	2	1.497	5	263
1.440	10	1.436	3	1.449	4	661
1.366	3	1.355	2	1.344	3	710
1.313	3	1.308	1	1.310	2	751
1.295	3	1.292	2	1.290	3	2.12.2

¹ Data reported by Suzanne van Dijke Beatty. The results reported for the other erroneously identified samples agree closely with these data and, therefore, are not repeated here. Some minor lines have been omitted from this table.

² Results obtained by K. Johansson, Vergleichende Untersuchungen an Anthophyllit, Grammatit, and Cummingtonit: *Zeit. Krist.*, **73A**, 43 (1930). Intensities reported in this instance rated from 5 for very strong to 1 for weak.

³ Fluor-amphibole prepared at the Electrotechnical Laboratory, U. S. Bureau of Mines in its current program on the synthesis of the asbestiform minerals. Chemical analysis of this aluminum-containing amphibole is as follows: SiO₂ 50.95%, Al₂O₃ 12.48%, MgO 19.95%, CaO 12.71%, and F 6.34%. This approximates the formula Ca₂(Mg₄Al)-(AlSi₇)O₂₂F₂. More detailed information on synthetic fluor-amphiboles will appear at a later date.

strongest lines in the chrysotile patterns, the (200) and (400) reflection, have d values of 7.3 ± 0.1 and 3.65 ± 0.03 Å.U.

THE NOMENCLATURE OF CLAY MINERALS

At the International Soil Science Congress held in Amsterdam (July 26–August 1, 1950) a special meeting to discuss the nomenclature of clay minerals was held which was attended by a representative number of workers in this subject. The following were discussed:

1. *ILLITE*.

Professor R. E. Grim emphasized the non-specific character of this term when it was first applied to mica-clay minerals. Following a discussion on the relation of such minerals to micaceous minerals containing randomly interstratified layers of other types, it was generally agreed that illite should be used as a general, non-specific, term for mica-clay minerals (both di- and tri-octahedral types, i.e., heptaphyllites and octaphyllites) which show no significant swelling characteristics and which give a first order, basal reflection of about 10 Å which is unaffected by mild chemical and/or thermal treatments.

HYDROUS MICA was suggested as a convenient general term to cover both illites and mixed-layer minerals such as bravaisite, hydrobiotite, etc.

2. *HALLOYSITE, METAHALLOYSITE, ENDELLITE*.

The confusion which exists in the use of these terms and which generally requires the use of additional explanatory words or phrases was discussed, and the general feeling of the meeting was that great simplification and clarification would result if the word 'halloysite' were employed for all forms of the mineral. When necessary, additional self-explanatory qualifications could be used such as: fully hydrated, partially hydrated, dehydrated, glycol-halloysite, halloysite to 7 Å, etc. . . .

It was unanimously recommended that the use of endellite should be discontinued.

After some discussion, it was also agreed, though less emphatically, to discontinue the use of metahalloysite; the view was expressed that the prefix 'meta' had no very clearly defined significance in mineralogy. The question has since been raised again in writing, firstly by Dr. D. M. C. MacEwan who, after looking into this question finds "numerous precedents for such a usage," and subsequently by Professor C. W. Correns who also finds many examples of meta-minerals which are dehydrated forms. Although these additional points cannot now be discussed by all who attended the meeting, it is probable that if they had been available, a majority of those present would have agreed to the continued use of metahalloysite as a convenient term for the largely dehydrated form which occurs naturally. It was emphasized by Dr. G. W. Brindley that the range of basal spacings exhibited by natural metahalloysites shows that they are rather variable materials. Therefore, even if metahalloysite is retained as a convenient term for naturally occurring materials, it does not remove the need for employing more precise qualifications when more precisely defined forms are under consideration.

3. *THE KAOLIN GROUP OF MINERALS*.

Dr. G. W. Brindley suggested that the group comprising kaolinite, dickite, nacrite, halloysite, etc., should be called the 'kaolin group' to avoid the confusion which may arise if the name of one member of the group (generally kaolinite) is also applied to the whole group. No confusion can arise with the use of kaolin as a rock name, since such a rock is simply one which is rich in kaolin minerals. This suggestion was received with general approval.

4. *MONTMORILLONOID GROUP*.

Dr. D. M. C. MacEwan pointed out that the use of montmorillonite in three different senses leads either to confusion or to an unwieldy qualification of the term. At present

montmorillonite is used for (a) the particular mineral from Montmorillon which is highly siliceous, (b) aluminous minerals generally of this type, and (c) all minerals of this type including nontronite, saponite, To avoid this confusion and also the lengthy term "minerals of the montmorillonite group" when the group as a whole is implied, the term "montmorillonoid" was suggested for this group of minerals. Montmorillonite can then be restricted to sense (b), with (a) as a particular case of (b).

There was considerable difference of opinion on this point and some adverse criticism on the ground that the termination 'oid' did not necessarily suggest a mineral akin to montmorillonite, c.f., chlorite and chloritoid. Professor C. W. Correns has suggested the term 'montmorin' for this group of minerals, which is similar to 'kaolin.' Montmorillonoid, however, has been employed in the Monograph "X-ray Identification and Structures of Clay Minerals" (Edited by G. W. Brindley, to be published by The Mineralogical Society), and whether or not it will be generally adopted remains to be seen.

G. W. Brindley (*Chairman*), and D. M. C. MacEwan (Great Britain), S. Caillère (France), C. W. Correns (Germany), J. Ch. L. Favejee (Holland), and R. E. Grim (U.S.A.).

ENROLLMENT OF GEOLOGISTS AND GEOPHYSICISTS IN THE NATIONAL SCIENTIFIC REGISTER

DAVID M. DELO

Planning for the mobilization and effective use of the scientists and engineers of the United States for purposes of national defense is now under way. Appointment of a committee composed of leaders in science and industry by the National Security Resources Board is a recent step. The representative of the geological professions on this committee is E. De Golyer; the chairman is Charles A. Thomas, vice-president of Monsanto Chemical Co. and former president of the American Chemical Society.

A basic element in the entire program for maximum use of the nation's scientific manpower will be an adequate scientific register. This will tell us the magnitude and nature of the national pool of technological competence.

In order to secure this information, the N.S.R.B. was instrumental last summer in causing the establishment of a National Scientific Register Office in the U. S. Office of Education. This office has let contracts with the American Chemical Society, the National Research Council and the American Institute of Physics for enrollment of all physical and biological scientists and mathematicians. Enrollment will be accomplished through the societies and the register will also be kept up to date through the societies. The American Geological Institute has been asked to ensure the enrollment of all geologists and geophysicists.

The project must start essentially from scratch because the records of the World War II National Roster are completely out of date. Also, with the tremendous increase in the number of working scientists since 1945, its records are very incomplete.

Some geologists and geophysicists who are included in American Men of Science, filled out a comprehensive registration form in 1948. These men *will not* be asked to answer all the questions on the new simpler form, but will be asked to fill out basic questions in the interest of uniformity.

The following methodology is being established for this project to ensure effective coverage. It is hoped that use of this method will spread the work, prevent wasteful and annoying duplication, and get the job done more promptly and effectively.

1. The register will include all geologists and geophysicists with a bachelors degree (or equivalent) and all graduate students.

2. Circularization will be accomplished by district committees. These will center in active local societies, state geological surveys and major departments.
3. Each committee will be responsible for full circularization in its district, and has been asked to estimate the number of forms required for this purpose.
4. Franked envelopes requiring no postage, will be utilized. Where possible, these will be pre-addressed.
5. Each form will be accompanied by a careful instruction memorandum.
6. A card file will be maintained in AGI headquarters showing which forms have been completed and certain other pertinent data. The register will be used for the following purposes—
 - (a) by Scientific Advisory Committees in the Selective Service System. It is expected that these committees will review appeals for deferment which are made to the President and that the information on file with the Register will be used in the consideration of such cases.
 - (b) by the National Security Resources Board. This agency is charged with responsibility for the planning of general mobilization both military and civilian, and will use the information in determining the requirements for scientific manpower in various activities.
 - (c) by the National Research Council and by the American Geological Institute in determining the distribution and availability of earth scientists among the various specialized occupations.
 - (d) as a means whereby Government agencies may identify personnel with specialized training or experience for unusual work requirements.
 - (e) in the preparation of studies and recommendations to Government agencies, such as the National Security Resources Board, concerning the needs for geological scientists in civilian occupations.

In addition, it will furnish information concerning the composition and structure of the geological professions which will be invaluable.

The cooperation of every member of the geological profession is earnestly requested so that our part in this national effort may be performed quickly and effectively.

The 25th Jubilee Meeting of the Swiss Mineralogical and Petrographical Society was held at Davos, Switzerland, on August 26–28, in connection with the annual assembly of the Swiss Natural History Society. During the first two days scientific sessions were held at which a number of papers were presented. The opening address, given by Professor Paul Niggli of Zürich, was entitled “Probleme der Alpine Metamorphose.” The third day was marked by a visit to the Institute for Snow and Avalanche Research on the summit of Weissfluhjoch and a jubilee luncheon there at which congratulations were extended to the society in French, German, English, Italian and Finnish. The presidential address was given by Dr. R. Galopin of Geneva. That evening at a general assembly of the Swiss Natural History Society, Dr. M. De Quervain, Director of the Weissfluhjoch Institute, spoke on “Metamorphose der Schnee.” Opening and closing dinners also were held together with the Natural History Society. About 120 persons attended the Jubilee Meeting. These represented 23 different countries. From the United States were present Wm. F. Bradley, Illinois State Geological Survey, A. E. Engel, California Institute of Technology, R. E. Grim, University of Illinois, and E. Wm. Heinrich, University of Michigan, delegate of the Mineralogical Society of American.

Two 8-day field excursions preceded the meetings and two more of similar length followed the assembly. All were well attended and unusually well organized and conducted.

Of particular interest to mineralogists was the later general mineralogical and petrographical excursion through the Alps which had as leaders P. Niggli, C. Burri, R. L. Parker, M. Vuagnat, E. Wenk, and E. Niggli.

E. WM. HEINRICH

During the 1950 general meeting of the German Mineralogical Society held in Göttingen during the end of August, a "Section for Crystal Knowledge," which was established by a group of interested scientists, was included within the scope of the Society. Professor H. O'Daniel, of the University of Frankfurt, who was elected director of the section, thus also becomes one of the two vice presidents of the German Mineralogical Society. In the "Section for Crystal Knowledge" the Society wishes to cultivate those subjects which extend beyond the borders of mineralogical problems alone and concern themselves with the growth and properties of all solid crystalline bodies. In this way the Society hopes to be placed in close contact with students of the allied sciences, mathematics, physics and chemistry. It is planned to hold an annual meeting for the presentation of papers as a means by which these contacts may be especially maintained. (*Translation by E. Wm. Heinrich from Deutsche Mineralogische Gesellschaft.*)

BOOK REVIEWS

DAS POLARISATIONSMIKROSKOP (Eine Einführung in die mikroskopische Untersuchungsmethodik durchsichtiger kristalliner Stoffe für Mineralogen, Petrographen, Chemiker und Naturwissenschaftler im Allgemeinen) by CONRAD BURRI, Professor at the Eidgenössischen Technischen Hochschule in Zürich, Switzerland, 1950, pp. 308, 168 text figures and 4 Plates. Verlag Birkhäuser, Basel, Switzerland.

This excellent book by Burri is a worthy successor to the *Traité de Technique Minéralogique et Pétrologique* published in 1907 by Duparc and Pearce of the University of Geneva, Switzerland. However, their treatise is more detailed with emphasis on the derivation of equations and the theoretical aspects of each problem than is the recent book by Dr. Burri. This difference is natural because Duparc and Pearce were in a University, while Professor Burri teaches in an Institute of Technology which is adapted to the needs of the engineer whose primary interest is in the applications and significance of an equation rather than in its derivation; in fact, many equations important to engineers are purely empirical and set up primarily to represent data of observation.

This distinction in attitude toward the subject is important and fundamental. Burri's presentation wisely seeks to familiarize the student with the methods of direct application rather than to cover the entire field. Even with this restriction the book contains 286 pages of reading matter. His descriptions of the many useful methods are clear and show good judgment in their selection. Citations to the literature are copious and are intended to serve the serious student who may wish to enter more deeply into the theoretical aspects of a problem. In these references to the literature Professor Burri quite naturally emphasizes the contributions by Swiss and German authors and overlooks some important papers by investigators of other lands.

The book is separated into Divisions *A* to *J* respectively; each Division is in turn divided into sections. In Division *A* the fundamentals of crystal optics are discussed; section I treats of the nature of light; section II, of the optical indicatrix and its applications. Division *B* describes the polarization microscope; section I treats of image formation by use of lenses; section II, of construction details of the polarization microscope, including discussion of polarizing and analyzing devices; section III describes certain accessories; in section IV useful hints are given on work with the microscope and on its treatment; in section V methods for preparing materials for examination under the microscope are described. Division *C* considers investigations in natural light. Division *D* on orthoscopic methods includes section I on interference phenomena (theoretical) and section II on practical applications; section III is on the measurement of path differences, section IV, on extinction angles, and section V, on optically active crystals. In Division *E* the phenomena of absorbing crystals (pleochroism) and methods for the exact measurement of the absorption of transmitted light are described. Division *F* is on conoscopic methods; in section II the technique of conoscopic observations are considered; in section III the characteristics of interference figures are discussed at length; in section IV the dispersion of biaxial crystals is analyzed; in section V conoscopic measurements are treated in detail while in section VI the conoscopic relations of optically active crystals are discussed. In Division *G* methods are discussed for the determination of refringence by means of the Becke line and of the Schrodter van der Kolk phenomenon for comparing the refractive index of a crystal with that of an immersion liquid; section II is on the application of the immersion method to isotropic and to anisotropic crystals and crystal sections. In Division *H* on the Fedorow and later methods based on the use of the universal stage: section I presents a general statement on the usefulness of these methods and on the factors which affect the degree of accuracy of the results obtained therewith. In section II practical applications of the method

are described; in section III special methods for use with the universal stage are included; section IV contains descriptions of conoscopic methods which may yield good results with the stage. In Division *J* the computation of extinction angles for random section and zones of biaxial crystals is sketched. Illustrative cases, such as the series of plagioclase feldspars, of olivines, and of orthorhombic augites, are described for which data on extinction angles are useful.

The book is remarkably free from typographical errors; the text figures are well drawn and easily legible and the half tone illustrations are well reproduced. On page 67 an error appears in the demonstration that, in a uniaxial crystal, the direction of the extraordinary ray OS for a wave whose normal ON is given, can be represented by the reciprocal of $p_1 Q_1$, in which Op_1 and OS are conjugate diameters, and that therefore the point Q_1 may be considered to represent the direction of the ray OS , its plane of vibration, and its velocity. This conclusion was clearly stated by Fletcher in his pamphlet on the Optical Indicatrix published in 1892.

The book is so clearly and competently written and so well illustrated that it will interest and help students of the polarization microscope and of the methods of its application to problems in a wide variety of fields.

FRED. E. WRIGHT,
Washington, D. C.

THE NININGER COLLECTION OF METEORITES, H. H. NININGER AND ADDIE D. NININGER.

The Nininger Collection of Meteorites, by H. H. Nininger and Addie D. Nininger, published by the American Museum of Winslow, Arizona, in 1950, is a bound book of 144 pages and 38 plates. This is not only a catalog of meteorites, it contains a brief account of the methods the authors followed in making this collection. These meteorites were acquired the hard way, through fieldwork, lectures and writing countless letters, and does not represent the work of individuals who have had the prestige of a well-known institution with its resources behind them. To fully appreciate this, this book and the accomplishments it represents, one needs to have had similar experience and to have known the authors.

This collection of 587 different meteorites contains 133 that are unstudied. It includes the main masses of 140 different meteorites. In most collections there are small specimens that actually have no value other than to increase the total number of meteorites represented. This collection has some of these, about 50 of the 587 meteorites are represented by specimens of less than five grams. Percentagewise this is about 9 per cent. To appraise this, the reader should compare the Nininger Catalog with the Ward-Coonley Catalog, published in 1904. This privately owned collection consisted of 603 different meteorites, but contained 112 meteorites of less than five grams, or about 18 per cent.

The Nininger Catalog lists the essential information expected of such publications, but a general overall appraisal indicates it was hurriedly prepared and without the advantage of critical suggestions from others. These authors, could and should, have given some important information in the section "Field Notes." The bulk of these comments are uninteresting and of questionable value. When this work is reviewed in the light of the years of the authors' field activities, it seems unfortunate they did not take the opportunity to give some more important data.

Many very fine cuts are included but some of the meteorites illustrated are so small they add nothing of value to the catalog. Several photographs seem to have little bearing on the subject. If it is the aim of a reviewer to find mistakes, an opportunity is presented: the Bennett County, South Dakota, iron is listed as a coarse octahedrite, while Plate 12

shows it is a hexahedrite; Albin, Wyoming, is listed as a stone, it is a pallasite, but the listing of errors is not the sole aim of this reviewer.

This catalog will be useful to everyone interested in meteorites and even though some of the material in the sections Introduction and Field Notes are of questionable value, this reviewer recommends this volume.

E. P. HENDERSON,
U. S. National Museum, Washington, D. C.

BULLETIN 156, MINERAL COMMODITIES OF CALIFORNIA, with accompanying map. Published in 1950 by the California State Division of Mines, Ferry Building, San Francisco, California. Price \$2.00; cloth-bound, 443 pages.

Without so planning, Bulletin 156 serves as a closing salute to the termination of California's Centennial celebration. It records a significant era in the mineral development of the State, and is most timely in appearance, as this is a period of marked industrial expansion, accompanied by an increasing demand for various minerals essential to national defense. The publication presents the geological occurrence, economic development, and utilization of California's mineral resources and provides, in a single volume, information and data especially prepared by a large number of staff members of the Division of Mines. The book is especially well documented with reference data, contains a detailed index, and is a reliable source reference that has definite appeal to mineralogists, geologists, mining and technical engineers, economists, and to the staff members of industrial and manufacturing firms.

Recognizing the fact that California geology is constantly being amplified by field study and research, the discovery and development of new mineral deposits is frequently an economic resultant thereof. The opening chapter discusses the natural environment and the geological aspects that affect the occurrence of California's mineral resources. It is an excellent summary of California geology from an economic aspect, and it likewise presents in some detail an outline of the State's geomorphic provinces, and illustrates the wide diversity of both geologic structure and mineral occurrence within the State. An excellent large-size insert map of California accompanies this chapter, indicating in various colors the distribution of mineral deposits, with a smaller map showing the geology and major rock units, together with the larger fault lines, particularly the San Andreas and the Garlock.

Subsequent chapters present a summary of mineral production, followed by detailed descriptions of the various mineral occurrences. These discussions are quite comprehensive, embracing generally the historical background, geological setting, production, and economic utilization. Much extremely useful and interesting information is afforded; for example, in the description of lignite coal the fact is disclosed that the United States is now self-sufficient in a new raw material, namely Montan wax, formerly imported from Germany, but now recovered in a plant processing the Ione lignite beds. A further item of interest to the reviewer appears under the heading of platinum, it being recorded that blister copper from the Iron Mountain smelter has carried some platinum. Such occurrence affords an interesting geological implication, as thus far all California platinum mining has been confined to placer deposits. The section upon silver discusses the famous old Calico district, the rich ores being principally cerargyrite with some embolite occurring as near-surface or epithermal deposits. Likewise described under the caption of Tungsten are the Atolia scheelite occurrences, the steeply-dipping fissure veins containing high-grade scheelite, from which the erosional products created the noted scheelite placer deposit named the "Spud Patch."

The foregoing in no degree exhausts the wealth of information, but rather, in the terms of the miner, be regarded as a grab sample. The bulletin is so fundamentally important that it should not be overlooked by those interested in any degree in California mineralogy and

geology, and by those who either utilize or are searching for natural mineral products to meet their commercial needs. A final word as to the term mineral commodities as included in the title, such expression being employed by the writers to define, without specific restriction, the useful constituents of the earth as are found within the State. This includes mineral fuels, industrial minerals, metals, quartz crystals, and virtually all else that is so vital to our economic and social life.

An important concluding section is a listing of mineral producers, dealers, and commercial laboratories. An exceptionally fine series of photographs accompany the text, which is further supplemented by a series of economic graphs, from which future trends might be predicted. Bulletin 156 will long serve as a reliable reference for California minerals, which, at the close of a century, are still being discovered.

J. B. NICHOLS,
Sacramento, California.

CONSERVATION OF NATURAL RESOURCES, edited by GUY-HAROLD SMITH. Published by John Wiley and Sons, Inc., New York, 1950, 552 pp., \$6.00.

This is a noteworthy symposium on problems and methods of conserving our national resources—public land, soil, forests and treecrops, grass, water, waterways and waterpower, wildlife and fish, minerals and mineral fuels, recreational facilities and manpower. There are 20 different contributors for the 23 chapters, including the editor, Guy-Harold Smith, of Ohio State University, who also presents a short preface. In the introductory chapter, The development of conservation in America, by Alfred J. Wright, also of Ohio State University, laudatory tribute is paid to, among others, C. R. Van Hise for his 1910 book, *The Conservation of Natural Resources in the United States*, which had an outstanding influence on the early conservation movement. Other well known geologic figures mentioned as having contributed importantly to the growth of conservation efforts are Major J. W. Powell, N. S. Shaler and Herbert Hoover.

Of interest to students of the earth sciences group are chapter 3 on soil groups, by L. A. Wolfganger of Michigan State College; chapter 4 on soil conservation by W. A. Rockie of the Soil Conservation Service; chapter 12 on water supply by J. H. Garland of Illinois University; chapter 16 on mineral conservation by the late W. M. Meyers of Pennsylvania State College; and chapter 17 on the mineral fuels by E. W. Miller, also of Pennsylvania State College.

The chapter on mineral conservation sketches the history of the mineral industries and discusses the nature of mineral resources and the factors influencing their estimation. Some recent estimates of United States mineral reserves are summarized. Possibilities for reduction of waste in extraction, in processing and in use, recovery of secondary minerals and use of substitutes also are explored. The chapter concludes with a section on national and international mineral policies.

The chapter on mineral fuels opens with a comparison of the relative importance of energy sources between 1800 and 1946. Coal is discussed under origin, rank, reserves, districts, production, uses and its conservation in mining, processing, consumption and conversion to heat energy. Next follows a review of the nature, occurrence, products, distribution and reserves of petroleum. Petroleum conservation hinges on developments in each of the industry's technical branches—exploration, drilling, production and refining. Natural gas and atomic energy also receive short sections in this chapter.

For a panoramic survey of conservation, its problems and practice, this work deserves recommendation even to those whose chief interests brush only lightly the more specialized aspects of this field.

E. WM. HEINRICH,
University of Michigan.

GEOCHEMISTRY by KALERVO RANKAMA and TH. G. SAHAMA. 912 pages, 54 figures, 211 tables. University of Chicago Press, 1950. \$15.00.

The first major book in any particular field must generally bear sharper scrutiny than its successors, and often can be expected to undergo more careful criticism. *Geochemistry*, by Rankama and Sahama, attempts in its own words "... to survey the broad field of geochemistry and to account for the present stage of geochemical knowledge. ..." If, throughout the course of his examination, the reader continues to recall that the authors intend the volume "... neither to serve as a textbook of geochemistry nor to give a complete account of all geochemical information," then he may well consider that the work achieves unqualified success in hitting its somewhat shotgun target.

The book may be considered to consist of five main sections: An introductory section; Part I. General Geochemistry; Part II. Manner of Occurrence of the Elements; seven appendixes; and a bibliography containing 735 entries. In their introductory remarks the authors attempt to define geochemistry and choose to follow the example of Goldschmidt who set the geochemical goals of not only establishing the terrestrial abundance relationships and distribution of elements but also of defining the laws governing these two factors. Thus the writers justify their complete omission of any petrological phase diagrams by inferring that the goal of the geochemist lies beyond, in the more fundamental task of delineating "... the properties of the elements or of their ions which are responsible for the enrichment phenomena observed." In this introductory part there is also a brief but interesting historical outline of geochemical study.

In Part I, which consists of 406 pages, are found chapters on meteorites; abundance of elements; terrestrial geochemical structure; distribution of elements among the geochemical subdivisions of the earth; a chapter each on the geochemistry of the lithosphere, hydrosphere, atmosphere, and biosphere; a comparison of geochemistry with cosmochemistry; and a chemical history of the earth. Chapter 5, the largest chapter of 170 pages, describes the geochemistry of the lithosphere and is the backbone of the book. Topics considered in this chapter are crystal chemistry, composition and chemical differentiation of magmas, formation of sediments and the exogenic cycle, and metamorphism.

Part II contains 36 short chapters, each describing an element or group of elements in terms of its manner of occurrence, its geochemical character, its minerals, its behaviour during magmatic differentiation and in the exogenic cycle, and its abundance. The appendixes are (1) the periodic system, (2) atomic weights (1948), (3) atomic and ionic radii, (4) electronic structure of elements, (5) geodetic and terrestrial data, (6) gravimetric conversion factors, and (7) ways of expressing concentration.

The overall plan and organization of the book are excellent, and particularly useful are the many tables and diagrams into which much critically evaluated material has been compressed. The type style is large and easy to read, and the literary style is clear and generally concise. Relatively few typographical errors were observed; e.g., p. 181 chrysober for chrysoberyl, p. 734 kylindrite for cylindrite, and Daly is listed in the bibliography under both Reginald Aldworth and Reginald A.

Some of the general defects of the work have already received comment by other reviewers: namely, the rather haphazard organization of the subject index and the lack of many important references not in English, French, or German. Papers in languages beyond these three may not be "... addressed to the international reader," but a survey worthy of its name cannot afford to disregard them. The writers follow what appears to be a widespread continental fondness for specialized and superfluous nomenclature, not only in general terms such as diadochic, reduzates, sedimentogenic, agpaitic, resistates, and magmagenic for which better known terms are in more widespread use, but also in mineralogical names under which appear such obscurities as oyamalite, mallardite, asbolan, nagatalite, manganomelane, and szmikite. In spite of a continuing emphasis on the quantitative distribution of elements, the authors allow themselves to become preoccupied with the enumeration of minutiae of their occurrence, often listing very rare species of hardly quali-

tative significance, whereas others of greater geochemical importance receive no mention, e.g., ludwigite under the boron minerals and the occurrence of V in aegirine.

The pegmatites and veins deserve less cavalier handling. No mention is made of the large body of information on pegmatite structures, which are of considerable value in deciphering the relations of the magmatic and hydrothermal stages in differentiation. The treatment of ore deposits includes assumptions of many minerals as pneumatolytic and omits any mention of the works of Lindgren, Emmons, or Graton on ore fluids, zoning, or classification of ore deposits. This section is regrettably provincial in emphasizing its European viewpoint. The discussion of diagenesis is elementary. On the other hand, the section on granitization, although favorable, is notably restrained in its treatment.

Occasionally a statement of obscure phraseology or dubious validity crops up, as on p. 342, "There is conclusive evidence that living bacteria may occur and be active in certain sediments and sedimentary rocks of considerable geological age, as high as Permian." Again on p. 462 one reads "... minerals of the garnet group are not found in igneous rocks with the exception of the pyrope-rich garnet of the eclogites." This assumes that all eclogites are igneous rocks, a doubtful case, and ignores melanite in feldspathoidal syenites, pyrope in peridotites, and spessartite in pegmatites. Moreover the statement is contradicted on p. 644 by the puzzler, "Spessartite occurs in aplitic granite pegmatites."

The number of these minor criticisms in no way detracts from the broad value of the work as a summary of geochemical processes and data. The book is characterized by an extraordinary usefulness that cannot be diminished by minor defects in its usability. It is an essential volume for all geologists and mineralogists who can afford it.

E. WM. HEINRICH,
University of Michigan.

INTRODUCTION TO THEORETICAL IGNEOUS PETROLOGY by ERNEST E. WAHLSTROM. John Wiley and Sons, Inc. New York. 1950. 365 pp., 138 figures, 31 tables. Price \$6.00.

This, the third of the recent books by Ernest E. Wahlstrom (*Optical Crystallography* and *Igneous Minerals and Rocks*), probably is the best of the group and is a worthwhile addition to the growing group of modern petrological textbooks. For it is a textbook, making no claim toward presentation of new data or original hypotheses. It is, however, a concise, well balanced and equitable digest of current viewpoints on magmas and their descendants.

The book is divided into two parts. Part I (102 pp.) includes four chapters: (1) Introduction, (2) Heterogeneous Equilibrium and the Phase Rule, (3) Equilibrium in Silicate Systems, and (4) Igneous Minerals. Chapters 2 and 3 form an excellent introduction for the beginning student to the comprehension of the phase rule and multiple component systems as related to igneous petrology. Chapter 4 presents brief and somewhat abrupt statements on the origins and petrogenetic significance of the plagioclases, potash-soda feldspars including perthites, feldspathoids, silica minerals, pyroxenes, amphiboles, olivines and micas.

In Part II are found the following chapters: (5) The Crust and Interior of the Earth, (6) Magmas—Source and Nature, (7) The Ascent and Emplacement of Magmas, (8) Crystallization of Magmas, (9) Igneous Differentiation, (10) Assimilation and Syntexis, (11) Granitization, (12) Late Magmatic and Postmagmatic Processes, (13) Origin of Rock Types, and (14) Classification of Igneous Rocks. The work concludes with an appendix, Physical-Chemical Concepts Useful to Petrologists, which is a poor man's short course in physical chemistry, and finds place in the book because the author believes that "... most American students of geology and mineralogy never advance beyond the introductory levels in mathematics, chemistry, and physics . . .," a doleful observation, whether fact or opinion.

The arrangement of the book presents some irregularities in the over-all concept. Chap-

ter 12 on deuteric and post magmatic processes could best follow as a consequence of igneous differentiation (Chapter 9). The final chapter on igneous rock classification is a dull millstone of questionable usefulness to the book and concludes with a puzzling section on the metamorphic facies concept. In Chapter 8, Crystallization of Magmas, the significance of many of the phase diagrams, whose mechanics are explained in detail in Chapter 3, might well have been employed.

In the introduction the author implies that magma and lava are identical or synonymous. It is regrettable that he does not list among the references at the end of the chapter on silicate systems the useful compendium by Hall and Insley (*Jour. Amer. Ceram. Soc.*, **16**(10), 1933).

One is impressed throughout the book by the valiant attempt to present the several sides to controversial subjects, yet this effort has, in some instances, led to noncritical summaries and will result in some confusion in the minds of average beginners who thus cannot distinguish among old historical chestnuts, untried neophytic notions, and tested concepts with sound factual and theoretical bases. An evaluation as critical as the coverage is complete would increase the value of the book.

The line drawings are well chosen and excellent throughout. The photographs vary in quality. Some are very good (Fig. 10, Chap. 12; Fig. 2, Chap. 4); others are poor (Fig. 1, Chap. 4; Fig. 3B and C, Chap. 11). None is worthy of being repeated: Fig. 5, Chap. 4 and Fig. 11, Chap. 12 are identical. The index is sufficiently complete.

The book fulfills its title adequately, for it will undoubtedly serve ably to introduce a multitude of geological students to the elements of petrogenetic theory. As such it belongs on the bookshelves of all teachers of petrology and recommends itself as a more than useful textbook in this field.

E. WM. HEINRICH,
University of Michigan.

ANNUAL MEETING

The thirty-second annual meeting of the Mineralogical Society of America will be held in Detroit, Michigan, on November 8–10, 1951, with headquarters at the Hotel Statler.

Abstracts of papers to be presented at the annual meeting must be received by the Secretary on or before *July 16, 1951*. Abstract blanks may be obtained from the Secretary.

C. S. HURLBUT, JR., *Secretary*

The spring meeting of the Society for Experimental Stress Analysis will be held at The National Bureau of Standards and The Wardman Park Hotel, Washington, D. C., May 16, 17, 18, 1951. All inquiries should be addressed to Dr. Edward Wenk, Jr., c/o The David Taylor Model Basin, Washington 7, D. C.

The 14th meeting of the Meteoritical Society will be held with the Pacific Division of the American Association for the Advancement of Science on Monday, Tuesday and Wednesday, June 18–20, 1951, at the University of Southern California. Titles and abstracts of papers to be presented at the meeting should be sent to John A. Russell, Secretary, Department of Astronomy, University of Southern California, Los Angeles 7, California.

NEW MINERAL NAMES

Lombaardite

H. J. NEL, C. A. STRAUSS AND FRANS E. WICKMAN, Lombaardite, a new mineral from the Zaaiplaats tin mine, central Transvaal: *Union S. Africa Dept. Mines, Geol. Survey, Mem.* **43**, 45-57 (1949).

CRYSTALLOGRAPHIC: Oscillation and Weissenberg photographs showed the mineral to be monoclinic, unit cell $a=16.10 \pm .05$, $b=5.60 \pm 0.1$, $c=18.20 \pm 0.1$ kX, beta 115.5° . The unit cell contains $\text{Ca}_{10}\text{Fe}_5^{+2}\text{Al}_{27}\text{Si}_{18}\text{O}_{89}(\text{OH})_5$. There is a pseudocell with dimensions $a/2$, b , $c/2$, beta 115.5° . Possible space groups Pm , P_2 , P_2/m . X-ray powder data are given. Goniometric measurements could not be made. Four equally well-developed faces were noted in the zone of the b axis, also cleavages were present in this zone.

CHEMICAL: Analysis by C. J. Liebenberg of a sample containing about 1% tourmaline gave SiO_2 31.88, Al_2O_3 35.85, Fe_2O_3 4.80, FeO 7.47, MgO 1.54, MnO 0.18, CaO 15.36, Na_2O 0.50, K_2O none, H_2O^+ 1.43, H_2O^- 0.04, B_2O_3 0.14, P_2O_5 , TiO_2 , and F trace; sum 99.19%. After deduction of tourmaline, this corresponds to $\text{Ca}_{10}(\text{Fe}, \text{Mg})_5(\text{Al}, \text{Fe})_{27}\text{Si}_{18}\text{O}_{89}(\text{OH})_5$.

PHYSICAL AND OPTICAL: Lombaardite occurs as dark brown needles, 0.01 to 0.06 mm. in diameter and up to 2 mm. long. Optically biaxial, positive, with $n_s(\text{Na}) \alpha = 1.756 \pm .001$, $\beta = 1.761 \pm .002$, $\gamma = 1.777 \pm .003$, $2V_\gamma$ (calcd.) 58° , $2V_\gamma$ (measured) $60 \pm 4^\circ$, $r > v$, strong. Pleochroism, X nearly colorless, Y amber-brown, Z tawny olive. Absorption $Y > Z > X$. Orientation $\beta = b$, optic axial plane parallel to (010). Sp. gr. = $3.85 \pm .02$.

OCCURRENCE: Lombaardite is a rare mineral, occurring in explosion breccia in a cassiterite-bearing pipe in granite in the Zaaiplaats tin mine, Potgietersrust district, Transvaal. The explosion breccia was composed of granite fragments with cavities containing tourmaline, calcite, quartz, fluorite, chlorite, pyrite, arsenopyrite, parisite, bastnaesite, and lombaardite, which coats all the other minerals. Lombaardite does not occur in unbrecciated normal tin-bearing parts of the pipe.

RELATIONSHIPS: Similar in composition to epidote and pumpellyite. It is noted that the pseudocell of lombaardite is somewhat similar to the unit cell of epidote. However, the much higher specific gravity of lombaardite indicates a different type of structure.

NAME: For Professor B. V. Lombaard, head of the Department of Geology, University of Pretoria.

MICHAEL FLEISCHER

Ktenasite

P. KOKKOROS, Ktenasit, ein Zink-Kupfersulfat aus Lavrion (Griechenland): *Tschermaks Mineralog. Petrog. Mitt.*, Ser. 3, Vol. **1**, No. 4, 342-346 (1950).

A specimen from the Kamaresa mine, Lavrion (also Laurion, Laurium), Greece, in the collection of the University of Thessaloniki, consists of porous smithsonite with a thin crust of glaucokerinite (basic sulfate of Cu, Zn, and Al). On this are aggregates of blue serpierite and rare groups of tabular blue-green crystals up to 1 mm. in size, of a new mineral. Goniometric measurements and Weissenberg photographs show it to be monoclinic, with beta $= 84^\circ 25'$ (Weissenberg), $84^\circ 36'$ (goniometric), and a_0 $11.16 \pm .02\text{\AA}$, b_0 $6.11 \pm .03$, c_0 $23.74 \pm .02\text{\AA}$. Probable space group $C_{2h}^2 - P2_1/c$. The forms $c(001)$, $a(100)$, $r(013)$, $q(101)$, and $m(201)$ were observed. X-ray powder data are given; they differ from those of serpierite.

The mineral is transparent, blue-green (no. 1010 of the Baumann-Prases color chart), luster vitreous. Hardness = $2-2\frac{1}{2}$ G. = 2.969. Optically negative, n_s alpha 1.511, beta $1.613 \pm .001$, gamma $1.623 \pm .001$, $2V$ $51 \pm 1^\circ$ (Universal Stage); beta and gamma by immersion method, alpha calculated; gamma = b .

Soluble in dilute acids and ammonia. Spectroscopic and microchemical tests showed only

Zn and Cu. Analysis on 2.5 mg. selected material gave ignition loss 19.50, SO_3 19.92, CuO 32.44, ZnO (by difference) 28.14%. This corresponds to $3(\text{Cu}, \text{Zn})\text{O} \cdot \text{SO}_3 \cdot 4\text{H}_2\text{O}$, or $(\text{Cu}, \text{Zn})_3(\text{SO}_4)(\text{OH})_4 \cdot 2\text{H}_2\text{O}$. With this formula and taking 8 formula weights in the unit cell above, G is calculated to be 3.18 (2.97 observed).

Named for Professor Const. A. Ktenas, Greek mineralogist, who died in 1935.

DISCUSSION: Further comparison with other basic copper sulfates such as langite would be desirable.

M. F.

Basaluminite, Hydrobasaluminite

S. E. HOLLINGWORTH AND F. A. BANNISTER, Basaluminite and hydrobasaluminite, two new minerals from Northamptonshire: *Mineralog. Mag.*, **29**, 1–17 (1950).

Detailed description; the preliminary note was abstracted in *Am. Mineral.*, **33**, 787 (1948).

M. F.

Kruzhanovskite

A. I. GINZBURG, Kruzhanovskite—a new mineral of the phosphate group. *Doklady Akad. Nauk S.S.S.R.*, **72**, 763–766 (1950).

CHEMICAL PROPERTIES: Analysis by O. A. Alekseev gave P_2O_5 35.30, Fe_2O_3 34.62, FeO none, MnO 16.39, MgO 1.30, CaO 1.50, Na_2O none, K_2O none, H_2O^+ 8.75, H_2O^- 0.95, insol. 0.56; sum 99.37%. This gives $\text{MnFe}_2(\text{PO}_4)_2(\text{OH})_2 \cdot \text{H}_2\text{O}$. A thermal analysis curve is given, which shows endothermal breaks at 210° , 380° , and 620° , and an exothermal break at 750° . The mineral melts at 990° .

PHYSICAL AND OPTICAL PROPERTIES: Monoclinic, in poorly formed crystals of prismatic habit, up to 2–3 cm. in diameter. No measurable crystals found. Cleavage basal, very perfect. Fracture uneven. Color brown to greenish-brown, bronze on cleavage face. Streak yellowish-brown. Luster vitreous to dull. Hardness $3\frac{1}{2}$ –4, sp. gr. 3.31.

Optically biaxial, positive, $2V$ 40 – 45° . Strongly pleochroic with α wine-yellow, β orange-brown, γ reddish-brown. Absorption $\alpha > \beta > \gamma$. Dispersion very great with $r < v$. Optic axial plane is perpendicular to (001). The indices of refraction are α 1.79 ± 0.005 , γ 1.82 ± 0.01 .

X-ray powder data are given; they differ from those of dufrenite, rockbridgeite, laubmannite, and beraunite.

OCCURRENCE: In the Kalbinsk pegmatite as the outer part of a large nodule of altered triphylite. Intimately associated with sicklerite, from which it probably formed.

NAME: For the Russian mineralogist, Vladimir Ilyitch Kruzhanovsky, one of the creators of the Mineralogical Museum of the Academy of Science, U.S.S.R.

M. F.

Scholzite

H. STRUNZ, Scholzit, ein neue Mineralart, *Fortschritte Mineralogie*, **27**, 31 (1948) (Publ. 1950) (abs.)

Platy and elongate crystals, millimeter in size, colorless to whitish-gray, have the composition $\text{Ca}_2\text{Zn}(\text{OH})_2(\text{PO}_4)_2 \cdot \text{H}_2\text{O}$. They are monoclinic, pseudo-orthorhombic, with a_0 6.46, b_0 9.05, c_0 7.55 Å., $\beta = 90^\circ$. The unit cell contains 2 molecules. Indices of refraction are $\alpha \sim 1.583$, $\beta \sim 1.589$, $\gamma \sim 1.604$, $2V$ small. Found in a feldspar pegmatite at Hagen-dorf near Pleystein, Oberpfalz, associated with sphalerite, triplite, feldspar, and quartz; these are primary minerals and scholzite is secondary. Named for Dr. A. Scholz, mineral collector and chemist of Regensburg, Germany.

DISCUSSION: Not to be confused with schultzite (=geocronite?), *Dana's System*, 7th Ed., Vol. I, p. 395.

M. F.

DISCREDITED MINERALS

Alumian (= Alunite)

HENRY BASSETT AND THOMAS HENRY GOODWIN, The basic aluminium sulphates: *J. Chem. Soc. (London)* **1949**, 2239-2279.

Alumian, supposedly $\text{Al}_2\text{O}_3 \cdot 2\text{SO}_3$, was described by Breithaupt in 1858. Two samples from the British Museum were examined; both were from the type locality, Sierra Almagrera, Spain, and one had a label in Breithaupt's writing. X-ray study showed that, as had been suggested previously by others, alumian is alunite.

M. F.

Brunckite

J. ZEMANN, "Brunckit"—kryptokristalline Zinkblende: *Tschermaks Mineralog. Petrog. Mitt.*, Ser. 3, Vol. 1, No. 4, 417-419 (1950).

An x-ray powder photograph of brunckite (supposedly amorphous ZnS) from the type locality (see *Am. Mineralog.*, **24**, 350 (1939)) gave a good powder photograph corresponding precisely to that of sphalerite.

M. F.

Histrixite

PAUL RAMDOHR, Der "Histrixit" der Curtin-Davis-Mine in North-Dundas, nordwest-Tasmania: *Neues Jahrb. Mineral.*, Monatsh. **1950**, No. 6, 139-141.

Histrixite was described in 1902 as a sulfosalt of composition perhaps $\text{Cu}_5\text{Fe}_3\text{Bi}_{14}\text{Sb}_4\text{S}_{32}$ (See *Dana's System*, 7th Ed., Part I, p. 469). Type material from the Australian National Museum, Sydney, was examined and found to be a complex mixture of at least ten minerals. Tetrahedrite, jamesonite, pyrite, bismuthinite, arsenopyrite, chalcopyrite, sphalerite, and marcasite were most abundant; traces of pyrrhotite, native Bi, bournonite, and an unidentified mineral were also noted.

M. F.

Arizonite (a mixture)

J. L. OVERHOLT, G. VAUX AND J. L. RODDA, *Am. Mineral.*, **35**, 117-119 (1950).

Randite (a mixture)

CLIFFORD FRONDEL, *Am. Mineral.*, **35**, 245-250 (1950).

Uranothallite (= Liebigite)

H. T. EVANS, JR. AND CLIFFORD FRONDEL, *Am. Mineral.*, **32**, 251-254 (1950).

Dihydrite, Ehlite, Lunnite, Phosphorocalcite, Prasine, Tagilite
(all = Pseudomalachite)

L. G. BERRY, *Am. Mineral.*, **35**, 365-385 (1950).

Alaskaite (a mixture)

R. M. THOMPSON, *Am. Mineral.*, **35**, 456-457 (1950).

Taylorite (= Ammonian apthitalite?)

CLIFFORD FRONDEL, *Am. Mineral.*, **35**, 596-598 (1950). Type taylorite has not yet been re-examined.

Arduinite (= Mordenite)

BRONSON STRINGHAM, *Am. Mineral.*, **35**, 601-604 (1950).

Lossenite (a mixture)

R. M. PEARL, *Am. Mineral.*, **35**, 1055-1059 (1950).

Louderbackite (= Roemerite)

R. M. PEARL, *loc. cit.*

Zepharovichite (= Wavellite)

R. M. PEARL, *loc. cit.*

Peganite (= Variscite)

R. M. PEARL, *loc. cit.*

Sphaerite (= Variscite?)

R. M. PEARL, *loc. cit.* Type sphaerite has not yet been re-examined.

M. F.

NEW DATA**Paratacamite**

CLIFFORD FRONDEL, On paratacamite and some related copper chlorides; *Mineralog. Mag.*, **29**, 34-45 (1950).

Paratacamite was originally described in 1806 by G. F. H. Smith as a rhombohedral dimorph of atacamite, $\text{Cu}_2(\text{OH})_3\text{Cl}$, but was stated by H. Ungemach in 1911 to be only twinned atacamite. It is now shown to be a valid species distinct from atacamite. Rhombohedral with unit cell, $a=13.65$, $c=13.95$ kX in hexagonal coordinates, a_{rh} 9.150 kX, alpha $96^\circ 28'$. An angle table is given. Optically, uniaxial positive without perceptible dichroism, $n_O=1.843$, 1.842 ; $n_E=1.849$, 1.848 , all $\pm .003$. Some grains are biaxial with $2V$ up to 50° , $G.=3.74$, 3.72 (measured), 3.75 (calcd.). A new analysis is given. Paratacamite is easily synthesized and occurs as one of the corrosion products of copper and brass exposed to salt-containing air and as an alteration product of nantokite (CuCl) and eriochalcite ($\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$).

M. F.

Botallackite

CLIFFORD FRONDEL, *loc. cit.*

Botallackite was described by A. H. Church in 1865 as $\text{Cu}_4(\text{OH})_6\text{Cl}_2 \cdot 3\text{H}_2\text{O}$, but has generally been considered to be atacamite. X-ray and optical data indicate it to be a distinct species. Qualitative tests and a spectrographic examination showed only Cu, Cl, and H_2O as major constituents. Optically biaxial, positive with $2V$ moderately large and strong dispersion, $r > v$. Indices of refraction, alpha $=1.772 \pm .003$, beta $=1.800 \pm .005$, gamma $=1.846 \pm .003$. X-ray powder data are given.

M. F.

Antofagastite = Eriochalcite

CLIFFORD FRONDEL, *loc. cit.*

Antofagastite, $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, was described by Palache and Foshag in 1938. It is shown to be identical with eriochalcite (not erythrochalcite) of A. Scacchi (1870). The name eriochalcite has priority.

M. F.